United States Court of Appeals

for the

District of Columbia Circuit

TRANSCRIPT OF RECORD

JOINT APPENDIX

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 19,275

51

WOLFGANG BAENITZ, and
CHEMISCHE WERKE WITTEN,

Appellants,

v.

DAVID M. LADD,
Commissioner of Patents,

Appellee

Appeal From the United States District Court
for the District of Columbia

United States Court of Appeals
for the District of Columbia Circuit

FILED JUN 11 1965

Nathan J. Paulson
CLERK

TABLE OF CONTENTS

Complaint, Letters Patent, filed June 27, 1963 1

Answer to Complaint, filed August 1, 1963 6

Excerpts From Transcript of Proceedings, June 1, 1964 8

Witnesses:

Mrs. Waltraude Stroschneider
- Direct 9
- Cross 16
- Redirect 17

Dr. Jacques Cyrus Morrell
- Direct 19
- Cross 24
- Redirect 26

Hans Schubert
- Direct 28

Opinion, Jackson, J., filed January 4, 1965 29

Order, Jackson, J., filed January 4, 1965 34

Motion to Vacate Judgment Under Rule 59 F.R.C.P., filed January 14, 1965 35

Order, Jackson, J., filed January 28, 1965 41

Notice of Appeal, filed February 26, 1965 42

Plaintiff's Exhibit No. 1, Specification 43

Plaintiff's Exhibit No. 1, Board of Appeals Decision 48

Plaintiff's Exhibit No. 2, table of chemical values 52

Plaintiff's Exhibits Nos. 3, 4, 6 and 7, diagrams , ... 52a

Defendant's Exhibit 1A, Christensen patent 2,022,494 53

Defendant's Exhibit 1-B, Feuge patent 2,745,749 56

Defendant's Exhibit 1-C, Kuhrt patent 2,634,278 60

Defendant's Exhibit 2, Bennett chemical dictionary 69

Defendant's Exhibit 3, Millard chemistry textbook 71

Defendant's Exhibit 4, Bailey, "Melting and Solidification of Fats" 74

Defendant's Exhibit 5, Bailey, "Industrial Oil and Fat Products" 76

DOCKET ENTRIES

1963	
June 27	Complaint
Aug. 1	Answer
1964	
June 1, 2	Hearing, Jackson, J.
1965	
Jan. 4	Post trial briefs
Jan. 4	Memorandum opinion, findings of fact, and conclusions of law, finding in favor of defendant against plaintiffs and dismissing complaint. Jackson, J.
Jan. 4	Judgment in favor of defendant against plaintiffs and dismissing complaint; costs against plaintiffs. Jackson, J.
Jan. 14	Motion of plaintiffs to vacate judgment under Rule 59 FRCP
Jan. 25	Opposition by defendant to motion of plaintiff under Rule 59
Jan. 28	Order denying motion of plaintiff to vacate judgment of Jan. 4. Jackson, J.
Feb. 26	Notice of appeal

JOINT APPENDIX

IN THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

Wolfgang Baenitz
Rheinische Strasse 105
Witten-Annen
Germany

and

Chemische Werke Witten
Gesellschaft mit beschraenkter Haftung
Witten/Ruhr
Germany

Co-Plaintiff

vs.

David L. Ladd
Commissioner of Patents
United States Patent Office
Washington 25, D. C.

Defendant

CIVIL ACTION
No. 1651-63

[Filed June 27, 1963]

COMPLAINT
LETTERS PATENT

1. This Civil Action to obtain Letters Patent arises under United States Code, Title 35, Section 145, as appears more fully hereinafter.

2. The co-plaintiffs are Wolfgang Baenitz, a citizen of Germany and resident of Witten-Annen, Germany, and Chemische Werke Witten, Gesellschaft mit beschraenkter Haftung, of Witten/Ruhr, Germany, a company incorporated under the laws of the Federal Republic of Ger-

many. Chemische Werke Witten is the assignee of the entire right, title and interest in and to the application for Letters Patent referred to hereinafter, of which Wolfgang Baenitz is the inventor.

3. The defendant, David L. Ladd, is the Commissioner of Patents of the United States and official resident of the District of Columbia and is sued as Commissioner of Patents of the United States.

4. The plaintiff, Wolfgang Baenitz, filed an application for patent in the United States Patent Office on February 12, 1959, entitled "Production of Fats", which was given the Serial No. 792,706.

5. Said application was filed in accordance with the laws of the United States and the rules of the United States Patent Office, and was duly prosecuted before the tribunals of the United States Patent Office in accordance with the laws of the United States and the rules of the United States Patent Office.

6. Said application was passed upon by the Primary Examiner, who refused to allow claims 25 through 30, which read as follows:

> 25. A method for controlling the melting point of hard and brittle fats within a relatively narrow desired range during the manufacture thereof by esterification of a fatty acid selected from the group consisting of saturated fatty acids having about 10 to 18 carbon atoms per molecule and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of controlling the melting point of the ester mixture produced by said esterification by selecting the quantity of said excess glycerol.
>
> 26. A method for controlling the melting point of hard and brittle fats within a relatively narrow desired range during the manufacture thereof by esterifying a fatty acid selected

from the group consisting of lauric, myristic, palmitic and stearic acids and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of controlling the melting point of the ester mixture produced in the esterification of said fatty acids by utilizing a selected predetermined quantity of said excess glycerol in said esterification.

27. A method for controlling the melting point within a relatively narrow predetermined range in the course of the manufacture of hard and brittle fats essentially consisting of esterifying a fatty acid selected from the group consisting of saturated fatty acids having from 10 to 18 carbon atoms per molecule and mixtures thereof with a predetermined excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of effectively and accurately controlling the melting point of said ester mixture by selecting the quantity of said excess glycerol used in esterifying said fatty acid.

28. A method for controlling the melting point of hard and brittle fats having a predetermined desired melting point, said hard and brittle fats being made by esterifying a fatty acid selected from the group consisting of lauric, myriatic, palmitic and stearic acids and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of effectively and accurately controlling the melting point of said ester mixture by selecting the quantity of said excess glycerol used in esterifying said fatty acid.

29. A method for controlling the melting point of hard and brittle fats with the desired melting point falling within a relatively narrow range, said hard and brittle fats being obtained by esterifying a fatty acid selected from the group consisting of saturated fatty acids having from 10 to 18 carbon atoms per molecule and mixtures thereof with an excess of glycerol in the presence of an esterification catalyst to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of directly controlling the melting point of the ester mixture produced by said esterification so as to fall within said relatively narrow range by effectively selecting the excess quantity glycerol used in the esterification of said fatty acid.

30. A method for directly controlling the melting point of hard and brittle fats having the desired melting point thereof falling within a relatively narrow range, said hard and brittle fats being obtained by esterifying a fatty acid selected from the group consisting of lauric, myristic, palmitic and stearic acids and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of directly establishing said desired melting point in the ester mixture produced by said esterification by utilizing a selected predetermined excess quantity of said glycerol in the esterification of said fatty acid.

7. Plaintiff, the applicant, appealed from the decision of the Primary Examiner to the Board of Appeals, which Board of Appeals, in a decision rendered on April 30, 1963, affirmed the rejection of the Primary Examiner whereby plaintiff was refused a patent.

8. The Commissioner of Patents, by the Board of Appeals, contends that claims 25 to 30 do not involve invention over United States patents No. 2,022,484 to Christensen, 2,634,278 to Kuhrt, and 2,745,749 to Feuge et al.

9. The applicant disagrees with the contentions of the Commissioner of Patents and contends that the method defined by claims 25 to 30 is a patentable invention over the teachings of the patents to Christensen, 2,022,494, Kuhrt, 2,634,378, and Feuge et al, 2,745,749, and that the rejection of claims 25 to 30 was improper.

10. The co-plaintiff further states that the said invention of claims 25 to 30 is new and useful, and was not known or used by others in this country before applicant's invention thereof, and not patented or described in any printed publication in this or any foreign country before applicant's invention thereof or more than one year prior to applicant's application for patent therefor, and not in public use or on sale in this country for more than one year prior to said application and not patented in any country foreign to the United States by applicant or his legal representatives on an application filed more than twelve months prior to said application for United States patent and not abandoned.

Wherefore, plaintiff prays that the Honorable Court decree that the Commissioner of Patents be directed to allow the said claims, and for such other and further relief as may be in order and required.

Paul M. Craig, Jr.
Attorney for Plaintiffs

[Filed August 1, 1963]

ANSWER TO COMPLAINT

To the Honorable the Judges of the United States District
Court for the District of Columbia

1. The defendant admits the allegations of paragraph 1 of the complaint.

2. The defendant admits that the co-plaintiffs are Wolfgang Baenitz, a citizen of Germany and a resident of Witten-Annen, Germany, and Chemische Werke Witten, of Witten-Ruhr, Germany. The defendant also admits that Chemische Wrke Witten is the assignee of the entire right, title and interest in and to the application for Letters Patent, Serial No. 792,706. The defendant asserts that, otherwise, he is without knowledge or information sufficient to form a belief as to the truth of the allegations of paragraph 2 of the complaint.

3, 4. The defendant admits the allegations of paragraphs 3 and 4 of the complaint.

5. The defendant denies the allegations of paragraph 5 of the complaint.

6, 7, 8. The defendant admits the allegations of paragraphs 6, 7, and 8 of the complaint.

9. The defendant denies the allegations of paragraph 9 of the complaint.

10. The defendant denies that the method of claims 25 to 30 is new and useful, and was not known or used by others in this country before applicant's invention thereof, and not patented or described in any printed publication in this or any foreign country before applicant's invention thereof or more than one year prior to applicant's application for patent therefor. The defendant asserts that, otherwise, he is without knowledge or information sufficient to form a belief as to the truth of the allegations of paragraph 10 of the complaint.

FURTHER ANSWERING, the defendant asserts that the plaintiffs are not entitled to a patent containing any of claims 25 through 30 of the application involved in this civil action, for the reasons given and in view of the references cited in the Examiner's action of May 8, 1961, the Examiner's answer, and the decision of the Board of Appeals in that application. Profert hereby is made of copies of the said action, answer, decision,and references.

Respectfully submitted,

/s/ C. W. Moore
Solicitor, United States Patent Office
Attorney for Defendant

July 30, 1963

I hereby certify that two copies of the foregoing ANSWER TO COMPLAINT were mailed today to Mr. Paul M. Craig, Jr., 915 Pennsylvania Building, Washington, D. C., attorney for the plaintiffs.

/s/ C. W. Moore
Solicitor

EXCERPTS FROM TRANSCRIPT OF PROCEEDINGS

Washington, D.C.,
Monday, June 1, 1964

The above-entitled matter came on for trial at 10:00 a.m. before the HONORABLE JOSEPH R. JACKSON, Judge, United States District Court.

[3]

EXHIBITS

* * *

DEFENDANT'S	MARKED	RECEIVED
No. 1 - Patent Office Ringed Folder with papers A-Christensen, B-Feuge Et Al., C-Kuhrt, D-Examiner's Action, E-Examiner's Answer and F-Decision of Board of Appeals.	12	12
No. 2 - Page 434 Concise Chemical and Technical Dictionary by H. Bennet.	108	123
No. 3 - Pages 93 & 94, Physical Chemistry for Colleges by E. B. Millard.	109	123
No. 4 - Page 48, Melting and Solidification of Fats, by Alton E. Bailey.	109	123
No. 5 - Page 48, Industrial Oil and Fat Products by Alton E. Bailey.	115	123

* * *

[14]

WOLFGANG BAENITZ,

a plaintiff herein, was called as a witness in his own behalf and, having been first duly sworn, was examined and testified as follows:

* * *

MRS. WALTRAUDE STROSCHNEIDER

[15] was called as an interpreter and was duly sworn.

(Note: All questions to the witness and answers to same were through the interpreter.)

DIRECT EXAMINATION

BY MR. CRAIG:

* * *

[16] Q. Before your invention described in the application, what was the customary procedure at Chemische Werke Witten to control the melting point? A. By changing the fatty acids by --

(The interpreter conferred with the witness briefly)

By variation of change of the fatty acid composition influence was taken upon -- on the melting point of the esterification product.

Q. Has there been any change since your invention concerning the method of producting the ester products?

* * *

[17] THE WITNESS: Yes, the method is being used.

BY MR. CRAIG:

Q. When you say "the method" what method do you refer to? A. With the method I mean the invention.

Q. Now, Mr. Baenitz, what are the advantages of the method of your invention? A. The advantages of the method of the invention are that while using under the use of the invention the melting point can be precisely arranged and predetermined with one and the same starting fatty acid.

Q. Is there any reason for utilizing fatty acids that have carbon atoms that number 12 to 18, and is there any reason for using as high as possible a number of carbon atoms in the fatty acids? A. There are reasons for utilizing the fatty acids having the carbon atom number of from 10 to 18 because they [18] occur in nature in seed fat.

* * *

[19] Q. Did you prepare this table from standard textbook information? A. The melting points have been taken from a standard handbook and also the molecular weights. The saponification and hydroxyl numbers have been calculated.

* * *

[20] Q. Referring to this chart, what does the left column mean? A. The carbon atom number of the fatty acids which are in the tri, di and mono-glycerides.

Q. What do the letters "MG" stand for? What is this designation? [21] A. That is the column for "Molecular Weight".

Q. What do the letters "VZ" designate? A. This is the "Saponification Number".

Q. What does the designation "SMP." mean? A. Melting point.

* * *

Q. The figures you have given, are they for the pure tri-glycerides, di-glycerides and mono-glycerides? A. Yes.

Q. These figures do not represent figures for any mixtures? A. No.

Q. To your knowledge, is it possible to obtain directly only the di or mono-glyceride by esterifying a fatty acid with glycerin?

* * *

[22] Q. By direct esterification with glycerin? A. No, not for Mono and Di; for Tri approximately.

Q. If you react a fatty acid with an excess amount of glycerin, do you get inherently Tri and Di-glycerides?

(The interpreter conferred briefly with the witness.)

Q. When esterifying a fatty acid with an excess amount of glycerin, do you get inherently Tri and Di or Tri, Di and Mono-glycerides? A. With an excess amount of glycerin I obtain Di, Tri and Mono in combination.

* * *

Q. Mr. Baenitz, I show you plaintiffs' exhibit 3 for identification. Can you tell us what it is? [23] A. This illustration represents a graph showing the esterification products which I have obtained. One fatty acid has been esterified with different amounts of excess glycerin.

Q. Did you prepare this graph? A. Yes.

Q. When did you make it? A. About three weeks ago.

Q. Is the data based on experiments made by you? A. Yes.

Q. Does this graph represent the various mixtures of mono-tri- and di-glycerides that you obtained with the same fatty acid mixture when you used different amount of glycerin? A. Yes, with different amounts of glycerin.

* * *

Q. Mr. Baenitz, looking at the chart marked plaintiffs' exhibit 3, can you tell us what the first left-column represents? [24] A. This is the analysis of an esterification product, of a fatty acid mixture with so much excess glycerol or glycerin as to obtain a hydroxyl number of 12.4. Also indicated in the table are the acid numbers, the saponification numbers and the melting point, as well as the percentage proportion of the free glycerin, free fatty acids, tri- di- and mono-glycerides.

Q. Looking at the various columns, does the chart indicate that in each of the tests that you have conducted going from left to right that you have used increasing quantities of glycerin for your esterification? A. Yes.

Q. Does the chart also indicate that you have not been able to obtain only tri-glycerides, only di-glycerides or only mono-glycerides? A. It is clearly evident from the illustration.

Q. If you are taking the example in the last right-column, if you use more glycerin what would happen? A. When using more glycerin, even a greater amount of glycerin, the ratio of mon- di- and tri-ester amounts would practically not change any further. Accordingly, monoester would no longer be formed.

Q. I think that is the wrong translation. No more monester would be formed. A. No more; yes. The free glycerin would stay as free [25] glycerin.

Q. In other words, if you take the last example, if you add free glycerin the ratios would remain the same between mon- di- and tri- and you would only obtain further larger quantities of free glycerin; is that correct? A. Yes.

* * *

[27] Q. When you esterify with glycerin, what is commonly referred to as the Stoichiometric amount of fatty acid? A. The Stoichiometric quantity for tri-glyceride means that for one mole of glycerin I have to use three moles of fatty acid.

Q. If you use less fatty acid does it mean that one or two of the hydroxyl groups remain without anything attaching onto them? A. When using less fatty acid I have more glycerin. In addition to the tri-glyceride I obtain partial glyceride, either mono- or di-; di- and mono-.

Q. Do you refer to the tri-glyceride as full esters? A. Yes.

Q. And do you refer to the di- and mono- as partial esters? A. Yes.

Q. And partial because some of the hydroxyl groups remain; is that correct? A. Yes.

* * *

[29] Q. Now, Mr. Baenitz, these various equations you have just demonstrated on the board, they are well-known, are they? A. Yes.

Q. They were well-known to you before you made your invention; is that correct? A. They have been known for a long time.

Q. I would like to refer again to the chart where we have passed from three H OOC R to two H OOC R and one. Now what we have discussed are the theoretical reactions, is that correct? [30] A. Yes.

Q. If in practice you proceed with a certain amount of excess glycerin to the fatty acid, what happens in practice, do you get a tri-di- or mono- pure, or what happens? A. When I have an excess amount of glycerin react with the fatty acid I obtain tri- di- and mono-ester; with a small glycerin excess, I have tri- and di-glyceride. And where the excess has obtained a certain value I obtain tri- di- and mono, I obtain an end product with a very large excess amount; always a mixture of mono- tri- and di- and free glycerin.

Q. Have you ever been able to perform any one, with the exception of the tri-glyceride, or approximately the tri-glyceride with the full esters, have you ever been able to obtain by simple esterification the di- or mono-glycerides? A. No.

Q. In other words, you cannot, if you esterify a fatty acid with glycerin, you cannot obtain mono- or di- directly, to your knowledge? A. Not pure.

* * *

[31] Q. What does plaintiffs' exhibit 4 represent? A. This graphical section shows the melting point of fatty acid esters in dependence upon the hydroxyl number.

Q. Were these graphs based on test data performed by you? A. Exclusively on data found experimentally.

* * *

[32] Q. Looking at the chart of plaintiffs' exhibit 4, Mr. Baenitz, you have a series of downwardly pointed curves, is that correct? A. Yes.

Q. This means that you do have a certain minimum of melting point temperature for a given hydroxyl number, is that correct? A. Yes.

Q. Now the melting point in each case of the graphs you have made -- I would like to refer to plaintiffs' exhibit 2. A. Yes.

Q. Can you tell us whether the minimum of the melting point in these curves lies below the corresponding melting points of the tri- di- and mono-glycerides? A. The melting points are below the melting points known for tri- di- and mono-glycerides.

Q. Does it mean that you have a mixture in your product of tri- and di- and that the product you obtain with the hydroxyl number of between 40 and 100 has a melting point which is lower than that of the pure tri- or di-? A. Yes.

[33] Q. Before your invention did you have any knowledge of this behavior? A. No.

Q. To your knowledge, was it ever known? A. No.

Q. Do you know whether this phenomenon has ever been used by others? A. Nothing is known of that.

Q. Are you familiar with the literature on fatty acids in esters? A. I think so.

Q. Have you ever found anything prior to your invention that would indicate that you are able to obtain a lower melting point as a result of direct esterification with the particular excess quantity of glycerin? A. I could take no such suggestion from the literature or from any other source.

Q. Do you, as a person having worked for a number of years, since '46, in this particular field, did you expect that there was such an effect whereby you could depress the melting point by esterifying with this excess amount of glycerin? A. I did not expect this prior to my investigation.

* * *

[37] Q. Now does this chart represent data based on actual tests? A. These are calculated values.

Q. In other words, this chart shows what the calculated amount of glycerin would be necessary per 1000 grams of fatty [38] acid to obtain mono-glyceride, di-glyceride and tri-glyceride? A. Yes.

Q. Does the blue line on the graph represent the theoretical quantity necessary for mono-glyceride? A. Yes.

Q. Does the green line represent the quantity necessary for di-glyceride? A. Yes.

Q. And does the red line represent the theoretical quantity necessary for tri-glyceride? A. Yes.

Q. Now in between the di- and tri-glyceride you have marked off four lines, and you have indicated them for hydorxyl numbers 40 to 100 and 60 to 80. What does this represent? A. This is the glycerin requirement for esters with hydroxyl numbers within the range of between 40 and 100. And the center lines are the range of 60 and 80 hydroxyl numbers.

Q. In other words, if you use a certain amount of excess glycerin which will give you a hydroxyl number of 40 to 100 you would fall between the two outer lines; is that correct? A. Yes.

* * *

[41] Q. Mr. Baenitz, I refer to the graph marked plaintiffs' exhibit number 7, and I will ask you does this represent a graph for various fatty acids plotting the hydroxyl number against melting point? A. Yes. In Centigrade.

Q. I refer to the various square marks in blue, red, black and green and an off-shade red. What do they represent? A. These are the melting points of the tri-glyceride which can be taken from the literature.

Q. In other words, the melting point of the tri-glyceride that you have plotted here for C 10, C 12, C 14, C 16 and C 18 are those indicated in the literature; is that correct? [42] A. Yes.

Q. Now for the tri-glycerides your hydroxyl number is zero, is that correct? A. Yes.

Q. The points you have marked in triangles, are they the melting points given in the literature for the di-esters? A. Yes.

Q. Now when you speak of di-esters they are pure di-esters? A. Yes, pure.

Q. And the points indicated in circles, are they the melting points indicated in the literature for the mono-esters? A. Yes.

Q. Now you have also indicated a straight line connecting the tri-to the di- to the mono-. If you had mixtures of tri- and di- and di- and mono-, would you expect that the melting points would behave according to the straight line? A. No. Before my invention I would think that the line would be straight.

Q. Now in each case you have also indicated a line which has a dip in the minimum and then rises again, the minimum in each case lying below the temperature of the lowest temperature indicated in the literature. Do you obtain this [43] minimum in the case of each of these fatty acids indicated? A. Yes.

Q. Does this minimum occur in the hydroxyl range of 40 to 100? A. Yes.

Q. It only occurs between 40 and 100, is that correct? A. Yes, the minimum falls into this range.

Q. Does a similar curve also obtain if you have mixtures of these fatty acids? A. Yes.

Q. And in each case are you able to obtain a melting point lower than the mono-glyceride? A. Yes.

Q. And anywhere in the literature has there been any mention or indication that you obtain the downwardly extended curve to the minimum? Prior to your invention. A. No, nothing was known to me of that.

* * *

[44] CROSS EXAMINATION

BY MR. NAKAMURA:

* * *

[45] Q. Mr. Baenitz, in the column under fatty acids, does 18 indicate an 18 carbon atom fatty acid?

(The Interpreter: I didn't understand the question. I am sorry.)

Q. Does 18 represent stearic acid? A. Yes.

* * *

[48] Q. Mr. Baenitz, would you refer to plaintiffs' exhibit number 6? Mr. Baenitz, do these lines for the mono-glyceride, di-glyceride and

tri-glyceride terminate at the left-hand side at C 18? A. For other fatty acids with a higher common atom number than 18 the curves extend further towards the left. We have here a section for fatty acids from C 18 to C 10.

* * *

[49] Q. Mr. Baenitz, would you say that from the position of the di-glyceride line for or at the value for the C 18 ester, if the excess of glycerin used is at the intersection between the C 18 line and the di-glyceride line, you would have an ester which would fall or have a hydroxyl number between 40 and 100? A. This relatively small range contains hydroxyl numbers within the range of from 40 to 100; precisely between 80 and 100.

* * *

[50] REDIRECT EXAMINATION

BY MR. CRAIG:

* * *

Q. If you esterify a quantity of fatty acid, and more particularly a quantity of stearic acid, with an excess quantity of glycerin to obtain a hydroxyl number of 40 to 100, isn't it true that your product is different from the pure di-glyceride? A. Yes.

Q. Now, if you use stearic acid with C 18 and you use an excess quantity to produce a hydroxyl number of 40 to 100, would you always get tri-glyceride, di-glyceride and mono-glyceride? [51] A. Yes.

* * *

Q. Let me rephrase the question. If you use stearic acid with C 18 and you use an excess quantity of glycerol to obtain a hydroxyl number of 90, what would you obtain in the mixture? A. Tri- and di-glyceride.

Q. Would you get a small amount of mono-? A. And a small amount of mono-, under 10 percent.

Q. Referring to plaintiffs' exhibit 6, where you have a small overlap between the green line of di-glyceride and the black line indicating the 100 line of hydroxyl, you could never get this alone, could you? You would always get tri-, di- and mono-glyceride; is that correct? A. Yes.

Q. Now your product that you obtain from direct esterification, that is different from a pure di-glyceride? A. Yes.

Q. And it is different because it is composed always of a mixture of tri- and di- and sometimes with some mono-; is that correct? [52] A. Yes.

Q. Now referring again to plaintiffs' exhibit 7, and to the graph representing the stearic acid, you have for the tri-glyceride a melting point of about 73 degrees Centigrade, is that correct? A. Yes.

Q. For the di-glyceride you have a melting point of about 77, is that correct? A. Exhibit 2 shows it precisely.

Q. Now in the range of between 40 and 100, what is the range of melting points that you obtain with your direct esterification? (The interpreter conversed with the witness briefly.) The melting point. What is the range of the melting point that you can obtain if you use an excess amount of glycerin to obtain a hydroxyl number of 40 to 100? What is the range of melting point you can obtain for your stearic mixtures? A. The melting point drops in the melting point of the tri-ester after the lowest point thereof from 10 to 15 degrees.

Q. Would you say that this is completely unexpected to have this drop? A. It is not to be foreseen, not to be anticipated.

Q. Is it because of this drop that you can use higher molecular carbons, higher carbon atoms in your mixtures to [53] obtain a lower melting point? A. Yes, in fatty acid because of their carbon atom number which would otherwise have a much too high melting point, I can obtain esters with the desired melting point with the aid of my invention.

Q. Can you accurately predetermine what the melting point will be for any fatty acid or fatty acid mixture with your invention? A. Yes.

Q. Can you tell it accurately beforehand? A. Yes.

Q. And does it involve any other steps than the usual esterification? A. No.

* * *

[54] DR. JACQUES CYRUS MORRELL

was called as a witness by and on behalf of the plaintiffs and, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION

BY MR. CRAIG:

* * *

Q. Now the equations given yesterday by Mr. Baenitz concerning the reactions to produce tri-, di- and mono-glycerides are only theoretical equations, isn't that correct? A. Yes.

Q. It doesn't necessarily follow that in reacting a fatty acid with glycerin you obtain only one or the other of the glycerides?[60] A. Not at all.

* * *

[61] Q. Dr. Morrell, do you understand what is meant by hydroxyl number? A. Yes, I do.

Q. Is this an indication of the free hydroxyl groups in the ester? A. Yes, it is.

Q. Is it difficult to determine the hydroxyl number, or is it well-known? A. It is well-known.

Q. There is nothing particularly difficult about it, is there? A. No. It is usually known as the acetyl number.

Q. This was known even twenty or thirty years ago, was it? [62] A. I would say at least forty because I became acquainted with it thirty-five years ago.

* * *

[68] Q. Dr. Morrell, if you were given the information from a textbook concerning the values of the melting point of the tri- di- and

mono-glycerides, would it be obvious to you, as an expert, that there is a decrease in melting point in the hydroxyl number of 40 to 100? A. Not at all.

* * *

[72] Q. Does this patent relate to a process of producing allegedly directly mono-glycerides or possibly di-glycerides? A. This is what the inventer claims here, either one or the other.

* * *

[73] Q. Now assuming that the patentee produces mono-glycerides and also possibly di-glycerides and mixtures thereof, would whatever mixture results from the process of this patent in the presence of a catalyst be the mixture that you obtain according to the process of claim 25? A. No, it could never be, because he uses excess glycerin.

Q. When you say excess glycerin, do you have reference to amounts of glycerin which are greater than that which is necessary for theoretically producing mono-glyceride? A. That is right.

Q. Do you mean excess glycerin remains free after the esterification reaction? A. According to my calculations, he has considerable glycerin left after the reaction.

* * *

[74] Q. If what Christensen teaches is true, namely the production of mono and di-glycerides, pure or in mixture, is there anything in this patent that would in any way teach that you can vary the melting point as a function of the hydroxyl number? A. He says nothing at all about melting point.

Q. Does he give any indication of melting point of his product anywhere? A. He doesn't mention it anywhere.

Q. Does he mention the range of 40 to 100 hydroxyl numbers? A. He doesn't mention the hydroxyl number.

Q. I would like, again, to call your attention to plaintiffs' exhibit 3, if I may, which is the chart prepared by Mr. Baenitz. If what the patentee Christensen discloses is true, and if what Mr. Baenitz discloses is true, would you say that whatever product you obtain by Christensen is completely different from what you obtain according to the teaching of the application involved in this suit? A. I would say, yes, it is completely different. It [75] would be more on the right-hand side of this chart.

Q. Well, it would mean that you would have, if what Christensen teaches, you would either have all blue, all red or mixtures of blue and red, but you would not have any green; is that correct? A. That is right, he claims all blue.

Q. All mono- and all di-? A. And all di-, either one or the other.

Q. But no tri-, he never mentions any tri-? A. No tri-.

Q. Now if you look at this graph, referring to the hydroxyl number, and you look at plaintiffs' exhibit number 3, and assuming they are correct, within the hydroxyl number of 40 to 100, would you not in the process according to the plaintiff, Baenitz, obtain a mixture that is predominant tri- and di-? A. Yes, according to this chart you would.

Q. Would you from that conclude that the novel unexpected results of your temperature drop is due to the fact that you obtain mixtures of tri- and di- with only minimum amounts of mono-glyceride, if any, present in the end product? A. I would say it must be because those curves represent the particular mixtures which are shown here with the hydroxyl numbers indicated, that is between 40 and 100.

[76] Q. Now did you also, Dr. Morrell, investigate the calculations made by the patentee and, in particular, did you investigate page 2, right column, lines 1 through 29, in which he sets forth a specific example of what he produces, and a statement that he can also produce di-glycerins or di-stearins or mixtures of mono- and di-? Did you investigate this in particular? A. Yes. I investigated this by checking the amount of glycerin that he used with the Stoichiometric

amount which was required for stearic acid. The amount of glycerin would be in the proportion of 92 to 284. He uses 120 parts of glycerin to 300 parts, and this shows an excess of approximately 14 percent of glycerin which, of course, he does not refer to. In fact, he does not even indicate that he has separated it. He states here that the product sets to an amorphous wax-like solid and on analysis shows a free fatty acid content of about 3.4 percent. He says: "This indicates that the reaction has proceeded to such an extent that the yield is about 96-97 percent of the theoretical for monstearin."

* * *

[78] Q. I will show you plaintiffs' exhibit 6, in which you have a curve of glycerin necessary against acid number. If what Christensen teaches, would you say that his product falls between the blue and the green lines at all times, mixtures of it? A. Yes, I would say so.

* * *

[79] Q. But as a whole, is there any indication in the patent, or any teaching or any suggestion that you have ascertained from your reading that would indicate that you can control the melting point within that particular hydroxyl range of 40 to 100 by direct esterification with the corresponding excess amount of glycerin? A. No, there is none whatever for two reasons. In the first place, there is a doubt whether he got it, but if he did get it, it would be on the high hydroxyl number side; if he got what he claimed.

* * *

[83] Q. Turning now to the Kuhrt patent, the patentee is concerned with separating out pure mono-glyceride from the reaction product which is a mixture of tri- di- and mono-; isn't that correct? A. That is right; yes.

* * *

[84] Q. Now looking at the Kuhrt disclosure of separating the mono- from the di- and tri- and also from the excess glycerin, is there any

indication in the patent that you can control by direct esterification the melting point of the final product by choosing a quantity of excess glycerin to give you a hydroxyl number of 40 to 100? A. No, there isn't, as far as I have been able to read. He doesn't even mention the melting point or the hydroxyl numbers.

* * *

[88] Dr. Morrell, if I may call your attention to column 1 of this patent and more particularly to lines 55 through 65, the patentee states: 'We have discovered that when a mixture of glycerides of long chain saturated fatty acids. . . " Now the glycerides of the long chain saturated fatty acids may possibly be the final product with which Baenitz is concerned, is that correct? A. That is correct.

Q. (Reading) ". . . containing substantial portions of long chain monoglycerides . . . " Since he says "substantial portions of long chain monoglycerides" it excludes the Baenitz products, doesn't it? [89] A. That is right.

Q. So even the glycerides that Feuge, the patentee, speaks about are different from the Baenitz products because Baentiz does not contain substantial portions of monoglycerides; is that correct? A. Yes. Monoglycerides are his starting material and he winds up with an entirely different compound.

* * *

[90] Q. Now I would like to call your attention to column 1, lines 69 and 70. He speaks about a hydroxyl number of 15 to 200, isn't that correct? A. Yes.

Q. Well, this is not the hydroxyl number of 40 to 100 that is referred to in the Baenitz application. It is a much wider range, in other words, isn't it? A. It is a wider range and it is the hydroxyl number of an acylated acetostearate compound rather than an ester of fatty acids.

* * *

[95] Q. Dr. Morrell, in summary, you have studied the Christensen patent, the Feuge patent and the Kuhrt patent, which are papers A, B and C in defendant's exhibit 1. Have you been able to find any indication in any of these patents that you can accurately control the melting point of the resulting ester mixture by choosing a predetermined amount of excess glycerin that you use during the esterification of the fatty acids? A. I have not been able to find the slightest indications (Note: Plaintiffs' Ex. No. 8 was received in evidence (Page 93)) [96] in these references.

Q. And given the information concerning the melting temperatures of the tri-, of the di- and of the mono-glycerides, would you say that it is unexpected or expected that the melting point of the resulting esterified mixtures lies below the tri-glycerides which has the lowest melting point? A. I would say that it is unexpected.

* * *

CROSS EXAMINATION

BY MR. NAKAMURA:

* * *

[102] Q. Now, Dr. Morrell, I would like to call your attention to lines 45 through 48 in that same column. Christensen states: ". . . and various mixtures of mono and distearin can be prepared by varying the quantity of stearic acid used, in other words by varying the ratio of glycerin to higher fatty acids." Now I would ask you this question: Suppose that an amount of stearic acid is used which is somwhat less than 2, let us say 1.95, times the amount needed to make mono-stearin. Now in the light of the last statement of Christensen to which I referred, would it be true that the ester product could be expected to contain more mono-stearin than the product formed when the amount of stearic acid is two times the amount needed to make mono-stearin? A. I would say that that would normally follow.

* * *

[103] (Reporter Reading: "Q. Now if the amount of stearic acid is further reduced, say to one and one-half times the amount needed to make mono-stearin, then would it be expected that the ester product would contain more mono-stearin, even more mono-stearin?")

THE WITNESS: Yes. I would say yes.

* * *

[112] Q. Dr. Morrell, I believe you have agreed that Christensen teaches that the amount of excess glycerin used would control the relative amount of mono- and distearin formed? A. Yes, I agree that various mixtures can be produced by varying the amounts of stearic acid and the ratio of glycerin to high fatty acids.

* * *

[114] (Reporter Reading: "Then, Dr. Morrell, directing your attention to the Christensen patent, the second column on page 2 in lines 42 through 45, where Christensen refers to the [115] preparation of distearin. He states that his product consists mostly of distearin. Would you say that Christensen there recognized the fact that you cannot get a pure distearin by esterification?")

THE WITNESS: Yes.

* * *

[123] Q. Now, Dr. Morrell, when we recessed I believe I was questioning you with reference to the material on the board; and I had asked a question whether it would be obvious that 1 gram of monostearin would theoretically contain about 3 and 1/2 times the number of unesterified hydroxyl groups that there are in 1 gram of distearin? A. Yes.

Q. Now, Dr. Morrell, would you say that it would be obvious that the hydroxyl number of a mixture of mono and distearin would depend upon theoretically the relative amount of mono and distearin formed? A. Yes, I would say so.

* * *

[126] Q. Then would it be clear from the statement that I just read out of Feuge et al. that the acylation of monostearin by Feuge et al, is esterification? A. Yes, it is esterification with acetic acid.

Q. Now, Dr. Morrell, further in column 1 of Feuge et al. in line 61, Feuge et al. state: ". . . while the mixture is unchanged in its chemical nature in that it is still a mixture of glycerides of saturated fatty acids, the mixture exhibits totally different properties . . ." Now focussing our attention particularly on the statement that "the mixture is unchanged in its chemical nature in that it is still a mixture of glycerides of saturated fatty acids", would it be obvious that the mixture or that the monostearin, after it has been acylated [127] is still an ester? A. Yes. The monostearin is not acylated and is still an ester. Was that your question?

Q. No. After acylation is it still an ester? A. Yes, it is an ester.

* * *

[130] REDIRECT EXAMINATION

BY MR. CRAIG:

* * *

Q. Now let us for the moment assume what what Christensen says is true and that he can achieve what he says. Does he teach the production of anything but mono- and di-glycerides? A. No, I do not see that.

* * *

[131] Q. I will read the definition of "hydroxyl number". (Reading) "The hydroxyl number is the number of milligrams of potassium hydroxide equivalent to the hydroxyl content of one gram of fat."

In other words, it is a correlation of the amount by weight of potassium hydroxide equivalent to the hydroxyl content in one gram of fat. Isn't it true that with fats of different molecular weight you would have different hydroxyl numbers? A. That is true, of course.

Q. Then your testimony on cross examination that the hydroxyl number only depends on the hydroxyl radical is not correct according to the definition even of defendant's [132] exhibit 5 in that the hydroxyl number also depends on the molecular weight of the fat in question? A. Yes, that is true.

Q. In other words, you have two factors that determine the hydroxyl number, one which is the radical, the OH group, and the other which is the molecular weight of the fat, because it is correlated always to one gram of fat regardless of what the molecular weight is; is that correct? A. That is correct.

* * *

[134] Q. Is there a difference between reacting two substances to produce a third and mixing two substances together which do not react? A. They are different because you can't always predict what you are going to produce.

THE COURT: Well, of course, when you have an admixture you have no chemical reaction, do you? That is the question; that is what he is asking you.

THE WITNESS: Yes, sir.

* * *

[135] Q. In other words, Feuge et al. start with a product as a starting product, namely the glycerides which are the end products in the Baenitz application; is that correct? A. Yes, that is correct.

Q. Would you speak up, please? A. Yes, that is correct. Either the end products of Baenitz or some other monoglycerides.

* * *

[136] Q. All right, let us go on. Acetic acid does not fall into the group of saturated fatty acids having carbon atoms from between 10 to 18, does it? A. No.

Q. It does not fall into that range? A. Not at all.

* * *

[140] MR. HANS SCHUBERT

was called as a witness by and on behalf of the plaintiffs and, having been first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION

BY MR. CRAIG:

* * *

[141] Q. Are you familiar at least from a management point of view with the production and sale of suppository masses made by Chemische Werke Witten? A. Yes, I am.

Q. Do you have information as to what was the process used before the discovery of Mr. Baenitz and what was the process used after and, if you do, could you tell the Court, please? A. Yes. I can say, in a few words, before the discovery of Mr. Baenitz it was necessary to produce some special esters from higher fatty acids up to 16 carbon atoms, and another ester made from higher fatty acids from 16 to 18 carbon atoms, and to mix these esters to get a special melting point. But it was very difficult to mix these products and to get a homogenous product. Therefore, suppository masses for market [142] production could not be produced to fill a variety of purposes. Pharmaceutical chemistry demanded some special melting points for the ingredients and through this invention of Mr. Baenitz it is possible to make suppository masses with melting points and you can put into these suppository masses whatever ingredients you want. You can put in peniciliin, et cetera.

Q. Is it necessary or does the industry require that the suppository masses your company makes be guaranteed to have a certain melting point? Is there any necessity for that? A. Yes. Unless we guarantee a special melting point the pharmaceutical industry will not buy our suppositories.

Q. You have to guarantee it? A. Yes. Otherwise we cannot sell our product.

Q. And by following the teachings in the application of Dr. Baenitz you are able to do it? A. We can do it.

Q. You can produce with a certain product which is based on natural fats a finished ester that you can guarantee to have a certain melting point; is that correct? A. Yes, we can guarantee it. It is very important. I can say that before the Baenitz discovery we manufactured suppository masses since 1947 and now have export in seventy countries of the world; and since the Baenitz invention the production has increased from 750 tons per year to 2,000. When you [143] consider a suppository only requires 2 grams of fatty base material you can calculate how much you can form from 2,000 tons.

Q. Do you have any competitors in this field of manufacturing suppository masses? A. Yes, we have. There are many competitors who make suppository masses, but they are unable to produce suppository masses up to now with the definite melting point such as we produce as a result of the discovery of Dr. Baenitz.

Q. Do you feel that your company has a definite competitive advantage over your competitors as a result of the discovery of Mr. Baenitz? A. Yes, we have.

Q. In your own experience and knowledge, do you feel that the invention of Mr. Baenitz has achieved considerable commercial success? A. Yes.

Q. Would you say that it is more than considerable commercial success? A. It is.

[Filed, January 4, 1965]

OPINION

This is an action pursuant to 35 U.S.C. 145, seeking a judgment of this Court authorizing the defendant, Commissioner of Patents, to issue Letters Patent of the United States to the plaintiffs containing claims 25 to 30 of an application for patent, Serial No. 792,706, filed February 12, 1959, entitled "Production of Fats."

The invention described in the application relates to a process for producing hard and brittle fats, suitable for food and pharmaceutical purposes, by esterifying a saturated fatty acid having between 10 and 18 carbon atoms per molecule, such as stearic acid, or a mixture of such acids, with an excess of glycerine in the presence of an esterification catalyst. The excess of glycerine is used to control the melting point of the resultant product. The application points out that the melting point depends upon the amount of excess glycerine used, and specifies the proportion of glycerine to be that amount which produces an ester product having a hydroxyl value of between 40 and 100.

Claim 25 is a representative claim, and reads as follows:

> A method for controlling the melting point of hard and brittle fats within a relatively narrow desired range during the manufacture thereof by esterification of a fatty acid selected from the group consisting of saturated fatty acids having about 10 to 18 carbon atoms per molecule and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, comprising the step of controlling the melting point of the ester mixture produced by said esterification by selecting the quantity of said excess glycerol.

The Patent Office Board of Appeals denied these claims on the ground that the difference between their subject matter and the prior art would have been obvious to one having ordinary skill in the art at the time the invention was made. 35 U.S.C. 103. To show this obviousness, the Board relied upon United States patents to Christensen (No. 2,022,494, granted November 26, 1935), Kuhrt (No. 2,634,278, granted April 7, 1953), and Feuge et al. (No. 2,745,749, granted May 15, 1956).

The Christensen patent discloses a process for producing edible fats by esterifying 120 parts by weight of glycerine with from 300 to

600 parts by weight of stearic acid in the presence of a catalyst. Christensen points out that a product containing 93% monostearin is obtained by using 300 parts by weight of stearic acid, and that a product "consisting mostly of distearin" can be obtained by using 600 parts by weight of stearic acid. He states, further, that ". . . various mixtures of mono and distearin can be prepared by varying the quantity of stearic acid used, in other words by varying the ratio of glycerin to higher fatty acids." The patent shows that 300 parts by weight of stearic acid is the quantity necessary to esterify one of the hydroxyl groups of glycerine, and that 600 parts by weight of stearic acid is the quantity necessary to esterify two of the hydroxyl groups of glycerine.

The Feuge et al. patent describes a process for modifying the melting point and like properties of monostearin by directly acylating a portion of the hydroxyl groups of monostearin. The process comprises acylating monostearin with 0.5, 1.0 and 2.0 moles of acetic anhydride per hydroxyl equivalent of monostearin to obtain various mixtures of monostearin and acetylated monostearin. Feuge et al. show that the amount of acetic anhydride used determines the hydroxyl value of the mixtures produced and controls the melting point of the mixtures.

The Kuhrt patent discloses the direct esterification of fatty material, such as stearic acid, with an excess of glycerine to produce ester mixtures containing a maximum amount of monoglyceride or a maximum amount diglyceride. Kuhrt recognizes that a partial ester product will be in dynamic equilibrium with glycerine and the fatty reactant, and that a change in the relative amounts of glycerine and fatty reactant will change the character of the product. To carry out the esterification as a cyclic process without reversion of the product, Kuhrt separates the partial ester from the reaction mixture by a rapid vacuum distillation.

At trial, the Patent Office introduced three additional publications.

The first was a textbook entitled "Physical Chemistry for Colleges", by E. B. Millard, First Edition, published in 1921, which disclosed on pages 93-94 that the melting point of a substance is lowered by the presence of some other substance in proportion to the quantity of it present.

The second, another textbook, "Melting and Solidifcation of Fats", by Alton E. Bailey, published in 1950, contained on page 48 the following statement:

> "It is well known that the melting point of a substance is usually lowered by the admixture of a small proportion of any second substance, even if the latter is relatively high melting."

The third was an article entitled "Industrial Oil and Fat Products", by Alton E. Bailey, published in 1945, which on page 48 defined "hydroxyl number" as follows:

> ". . . The hydroxyl number is the number of milligrams of potassium hydroxide equivalent to the hydroxyl content of one gram of fat." (Emphasis quoted).

The theory of the Patent Office was that either the Christensen disclosure or the Kuhrt disclosure, when viewed with the Feuge et al. patent in mind, would have made it obvious to a person skilled in the art to arrive at the subject matter of plaintiffs' claims.

The plaintiffs made three principal assertions: first, that Christensen did not specify the hydroxyl number for his products, or state that he sought to directly control their melting point; second, that Kuhrt did not suggest plaintiffs' method for directly controlling the melting point of the product, and third, that Feuge et al. was concerned with products having properties entirely different from the properties of the products obtained by plaintiffs.

The Patent Office responded by observing, first, that Christensen, through the use of the process and ingredients he disclosed, would inherently arrive at plaintiffs' results. Second, the Patent Office stated that although Kuhrt did not mention melting points, he did show that it was old to produce partial ester mixtures (containing various amounts of monostearin or distearin) by esterifying directly an excess of glycerine with stearic acid. Third, the Patent Office pointed out that the importance of the Feuge et al. patent resided in its disclosure that through varying the relative amounts of monostearin and acetic anhydride used in the direct acylation of monostearin, the melting point of the ester produced could thereby be controlled. Finally, the Patent Office relied upon the publications introduced at trial to show the amount of novelty involved in controlling the melting point of mixtures through proportionate variation of their ingredients.

After examining the evidence as a whole, the Court is inclined to agree with the defendant's interpretation of the references. The testimony demonstrated that the amount of glycerine used in the plaintiffs' process ranges from 131 to 168 grams per 1000 grams of stearic acid, and that this clearly overlaps the Christensen range of 160 to 168 parts. Further, it was shown that the hydroxyl numbers of esters obtained from use of 160 to 168 parts of glycerine would be in the range from 90 to 100. It was also pointed out that the temperature at which the Christensen process is conducted is the same as that for the plaintiffs' process. From this, it appears that the processes of Christensen and the plaintiffs are essentially the same. Accordingly, it seems reasonable to suppose they would inherently produce the same result. At least, the Court cannot say such a view is unwarranted where plaintiffs have offered no convincing proof to the contrary.

On the question of controlling the melting point, it is clear, first, that if Christensen's process inherently produces plaintiffs' result, then the feature of controlling the melting point would necessarily be included in such a process. Whether this advantage in fact occurred to Christensen appears somewhat doubtful. It is clear, however, from the other references, that varying the proportion of ingredients in mixtures such as Christensen's was, at the time of the invention, known to vary the mixture's melting point. This was shown by the various textual publications introduced by the defendant at trial, and also by the patent to Feuge et al. In view of this, the Court cannot say the Patent Office was clearly wrong in finding that control of the melting point as claimed by plaintiffs, would have been obvious to a person skilled in the art having all these disclosures before him.

Since the Court agrees with the Patent Office rejection on the above discussed theory, it does not find it necessary to reach the rejection based upon the Kuhrt disclosure.

The Court finds for the defendant, and against the plaintiff, and will dismiss the Complaint.

The above Opinion contains Findings of Fact and Conclusions of Law.

Dated: January 4, 1965.

JOSEPH R. JACKSON
United States District Judge

[Filed, Jan. 4, 1965]

ORDER

This cause came on for trial on June 2, 1964. Upon consideration of the record herein, as well as the briefs which the Court accorded counsel for plaintiffs and defendant an opportunity to file, it is this 4th day of January, 1965,

ORDERED, that judgment be, and the same is hereby entered in favor of defendant, and that the Complaint be, and is hereby dismissed, with costs to be assessed against plaintiffs.

JOSEPH R. JACKSON
United States District Judge

[Filed, Jan. 14, 1965]

MOTION TO VACATE JUDGMENT UNDER RULE 59 F.R.C.P.

Plaintiffs respectfully move to vacate the judgment of January 4, 1964 in the above cause of action since the principal underlying reason appears to be based on a glaring error. More particularly, on page 6 of the opinion, one finds the following language:

> The testimony demonstrated that the amount of glycerin used in plaintiffs' process ranges from 131 to 168 grams per 1000 grams of stearic acid, and that this clearly overlaps the Christensen range of 160 to 168 parts. (Emphasis added).

Turning to the Christensen patent, this patent utilizes 120 parts by weight of glycerin for 300 parts by weight of stearic acid (page 2, right column, line 1 through 16) to produce a substantially pure monostearin.

Likewise, to obtain substantially pure distearin, Christensen suggests doubling the quantity of stearic acid, that is 600 parts by weight of stearic acid for 120 parts by weight of glycerin.

In other words, the Christensen disclosure teaches the use of 300 to 600 parts by weight of stearic acid for 120 grams of glycerin.

Plaintiff at trial indicated that he utilizes 131 to 168 grams of glycerin per 1,000 grams of stearic acid to produce a product having a hydroxyl number falling within the range of 40 to 100. Hence, by simple calculations, the disclosure by Christensen teaches the following amounts of glycerin per 1,000 grams of stearic acid:

To produce substantially pure monoglyceride with 1,000 grams of stearic acid:

$$\frac{1,000}{300} \times 120 = 400$$

To produce substantially pure diglyceride with 1,000 grams of stearic acid:

$$\frac{1,000}{600} \times 120 = 200$$

Hence, whereas plaintiffs utilized 131 to 168 grams of glycerin per 1,000 grams of stearic acid, Christensen utilizes 200 to 400 grams of glycerin per 1,000 grams of stearic acid to produce diglyceride, monoglyceride, and mixtures of di and monoglycerides.

Hence, the range of 131 to 168 grams per 1,000 grams of stearic acid as used by plaintiff does not overlap the Christensen range of 200 to 400 grams of glycerin per 1,000 grams of stearic acid.

The opinion then continues on page 6:

> Further, it was shown that the hydroxyl numbers of esters obtained from use of 160 to 168 parts of glycerin would be in the range from 90 to 100.

Assuming that this is the case in connection with plaintiffs' process, it does not follow, as further stated on page 6 of the opinion

> "that the processes of Christensen and the plaintiff are essentially the same. Accordingly, it seems reasonable to suppose they would inherently produce the same result."

The attention of the Court is respectfully directed again to plaintiffs' reply brief on page 8 where the same calculations as made above were submitted to point out the absence of overlap between the range of 160 to 168 parts by weight of glycerin as used by plaintiffs with the range of 200 to 400 parts by Christensen. As a matter of fact, the absence of such overlap was acknowledged by the defendant in the brief for the defendant on page 8 where the defendant urged that Christensen, though totally silent, should be presumed to have used 80% crude glycerin. However, in the absence of any indication, it would indeed be strange to assume that plaintiffs use 100% and

Christensen, also silent, would use only 80% pure glycerin. Furthermore, pages 526 and 527 of the Condensed Chemical Dictionary, attached to the plaintiffs' reply brief, clearly dispel any notion that anything but pure glycerin would be used normally by both plaintiffs and Christensen in view of the nature of intended use of their respective products in the food and pharmaceutical industries.

Though Christensen does not indicate any hydroxyl numbers for his product, it was pointed out at trial that pure distearin has a hydroxyl number of 89.8 and pure monostearin a hydroxyl number of 313 (plaintiffs' exhibit 2). Assuming the process of Christensen operates as disclosed by the patentee, his products consisting of pure distearin as well as a mixture essentially consisting of pure distearin with small quantities of pure monostearin might have a hydroxyl number of 89.8 to 100. However, in that case, the product would be completely different from that of plaintiffs whose product consists of mixtures of tristearin, distearin, and monostearin when esterifying with an amount of excess glycerin to produce an end product having a hydroxyl number of 40 to 100 (plaintiffs' exhibits 2, 3, and 6).

In other words, whereas plaintiffs have established at trial that the use of 131 to 168 grams of glycerin per 1,000 grams of stearic acid produces a product having a hydroxyl number of 40 to 100 and inherently always consisting of mixtures of tristearin, distearin, and monostearin, only two alternatives exist insofar as Christensen is concerned leading to the following conclusions:

(1) By using 200 to 400 grams of glycerin per 1,000 grams of stearic acid, Christensen produces pure monostearin by using 200 grams of stearic acid, pure distearin by using 400 grams of stearic acid and mixtures of pure mono and distearin by using 200 to 400 grams of glycerin per 1,000 grams of stearic acid. In that case, with products consisting of pure distearin or mainly pure distearin and small amounts of monostearin, Christensen would obtain a hydroxyl

number of 89.8 to 100 (overlapping the claimed hydroxyl number range of plaintiffs) but his products of pure diglyceride or mixtures of pure diglyceride and pure monoglyceride would be completely different from plaintiffs' products consisting at all times of mixtures of triglycerides, diglycerides, and monoglycerides. For example, plaintiffs' exhibit 2 indicates that the melting point of pure distearin is about 78°, of pure monostearin 81.5°, whereas pure tristearin has a melting point of 72°. There is no indication that within the range of hydroxyl numbers of 40 to 100 the unexpected results of temperature control would be achieved if the product consisted of pure distearin and monostearin. Additionally, since the end products of Christensen under this assumption are different from those of plaintiffs, the process cannot be the same.

(2) Assuming that Christensen's teachings are wrong as to the production of pure monostearin and pure distearin (which appears to be borne out by the Kuhrt patent 2,734,278) and assuming that Christensen's process would produce the same products as plaintiffs process, then it is obvious that the final product of Christensen could not have a hydroxyl number within the range of 40 to 100 since the amount of glycerin used by Christensen, that is 200 to 400 grams per 1,000 grams of stearic acid, would produce a final product that has a hydroxyl number greatly in excess of 100. By interpolating plaintiffs' exhibit 6, the hydroxyl number of Christensen, if his process were identifical would be about 150.

The foregoing analysis would indicate that either (1) Christensen has a different process producing different products from those of plaintiffs in which case an overlap in the hydroxyl number range with plaintiffs' claimed range would occur or (2) if Christensen's process were identical with that of plaintiffs, the products obtained by Christensen would be products having a hydroxyl number far in excess of the maximum number of the claimed range.

At trial plaintiff Baenitz testified that he was never able to obtain pure diglycerides or pure monoglycerides by direct esterification (R-30) which would seem to indicate that the alternative (2) would be the correct one, also borne out by the existence of the Kuhrt patent 2,634,278, which is presently practiced commercial process developed by Eastman Kodak Company to convert mixtures of tri-, di-, monoglycerides into pure monoglycerides or pure diglycerides.

Since the defendant has attempted to becloud the issues an attempt will be made below to restate the issues before the Court in order that a decision can be obtained based on proper technical premises.

1. It is well known to esterify fatty acids having 10 to 18 carbon atoms per molecule by reacting the same with glycerin.

2. It is also well known that this reaction inherently produces mixtures of triglycerides, diglycerides, and monoglycerides. That it is practically impossible to obtain pure diglycerides and pure monoglycerides by direct esterification is borne out by the Kuhrt patent 2,634,278 which utilizes a complicated process for converting the mixtures constituting the end products of plaintiffs' process into pure monoglycerides or pure diglycerides.

3. It is also known that pure triglycerides, pure diglycerides, and pure monoglycerides have different melting points so that mixtures thereof in different proportions of the tri-, di-, and monoglycerides can be assumed to have different melting points.

4. The sole issue presented by this case is whether it was obvious to directly control the melting point during the esterification step itself of the fatty acid by the use of an amount of glycerin producing a hydroxyl number of 40 to 100 in the final product resulting therefrom, whereby within this range an unusual temperature control exists which causes the melting point to dip (plaintiff's exhibit No. 7).

To place defendant's exhibits No. 1-B and 1-C in the proper perspective insofar as their real significance is concerned in the art, it is noted that U. S. patent 2,634,278 to Kuhrt (defendant's exhibit No. 1-C) utilizes a similar reaction as used in plaintiffs' process by reacting the fatty oil with glycerin and a catalyst, thereby producing mixtures of tri-, di-, and monoglycerides. However, the only concern of this patent is to convert the final product such as obtained by plaintiffs' process into substantially pure monoglyceride or pure diglyceride. In other words, this patent teaches a process utilizing as starting material in its process the final products obtained by the process of plaintiffs. Temperature control is of no concern to this patentee.

The Feuge et al patent 2,745,749 (defendant's exhibit 1-B) is concerned with a process for chemically modifying certain glyceridic mixtures (Column 1, lines 21 through 23) and utilizes as starting material the monoglycerides, i.e., the products obtained by the Kuhrt process. The chemical modification of the glycerides results in obtaining new products which have "unique value in food and drug applications" (Column 1, line 43), and, of the changes in properties, those of texture and flexibility are sought after while changes in melting point are merely incidental in this patent and were set forth only to identify the resulting products.

Hence, in view of the foregoing, it appears that the basic issue in this case whether patentability exists in the new and useful process or new and useful improvement in directly controlling the melting point as specified in the claims bearing in mind the definition under 35 U.S.C. 100(b) which defines the term "process" as follows:

> (b) The term "process" means process, art or method and includes a new use of a known process, machine, manufacture, composition of matter, or material.

In view of the fact that the judgment appears to be based on the obvious errors from a technical point of view, pointed out above, it is respectfully requested that the judgment for defendant be vacated and judgment for plaintiffs be entered.

To the extent that the Court may wish a rehearing, this motion also respectfully requests such rehearing.

Respectfully submitted,
DICKE & CRAIG
Attorneys for Plaintiffs

[Certificate of Service,
January 14, 1965]

By /s/ Paul M. Craig, Jr.

[Filed, Jan. 28, 1965]

ORDER

The Court having considered plaintiffs' motion to vacate the judgment rendered January 4, 1965, together with the defendant's opposition thereto,

AND the Court having found that a technical inaccuracy appears in the Court's opinion,

AND the Court having found that the said inaccuracy is insufficient, in view of the evidence as a whole, to alter the Court's ultimate determination.

IT IS, this 28th day of January, 1965,

ORDERED, that the Opinion, page 6, line 11, be amended by striking out "168 parts" and substituting therefor the following:

> ". . . 320 grams per 1000 grams, assuming that 80% crude glycerine was used in the reaction".

AND IT FURTHER ORDERED, that plaintiffs' motion to vacate the judgment of January 4, 1965, be and hereby is denied.

JOSEPH R. JACKSON
United States District Judge

[Filed Feb. 26, 1965]

NOTICE OF APPEAL

Notice is hereby given that Wolfgang Baenitz and Chemische Werke Witten, plaintiffs above named, hereby appeal to the United States Court of Appeals for the District of Columbia Circuit from the Judgment entered in this action on January 4, 1965, as amended in the Order denying Plaintiffs' Motion to Vacate the Judgment under Rule 59 F.R.C.P., the Order having been entered in this action on January 28, 1965.

Dated: February 26, 1965

DICKE AND CRAIG

By Donald R. Antonelli

Attorneys for Appellants

[Certificate of Service]

[Plaintiff's Exhibit No. 1]
[page 1]

D/3051

IN THE UNITED STATES PATENT OFFICE

Inventor:	WOLFGANG BAENITZ
Invention:	PRODUCTION OF FATS

Dicke, Craig & Freudenberg
(Registration No. 17,394)
915 Pennsylvania Building
425 Thirteenth Street, N. W.
Washington 4, D. C.

SPECIFICATION

To all whom it may concern:

Be it known that I, WOLFGANG BAENITZ, a citizen of Germany, residing at Rheinische Strasse 105, Witten-Annen, Germany, have invented certain new and useful improvements in the

PRODUCTION OF FATS

of which the following is a specification.

[page 2]

This invention relates to the production of brittle and hard fats. More particularly, the present invention relates to the production of brittle and hard fats from saturated fatty acids having between about 10 and 18 carbon atoms per molecule and glycerol.

Brittle and hard fats are required for a variety of purposes in e.g. the pharmaceutical and food industries. With the exception of cocoa butter, none of the known natural fats possess these characteristics while [page 3] the so-called modified fats or synthetic fats similarly fall short of required standards. It is well known to effect hardening of fats by catalytic hydrogenation but the hydrogenated products, e.g.

hydrogenated cocoa fat or palm kernel fat, have an increased melting point of about 35°C. but do not possess the desired hardness and brittleness.

It is known that fats having the desired chracteristics hereinbefore mentioned can be obtained by subjecting a free fatty acid derived from a natural fat or oil to hydrogenation and to esterification with a polyhydric alcohol, such as glycerol; these steps being effected in either order. In such known methods, the free fatty acid is preferably employed in the form of a vegetable fat or oil which melts in the region of that desired in the final product and which has been split in the usual manner. The products obtained by the direct hydrogenation of the initial fat or oil are not the same as, indeed, are markedly softer and less brittle than the products obtained by the above discussed prior art method. By this known method the particular melting point may be easily determined by a particular selection of the fatty acids that are to be reacted.

It has now been discovered that improved brittle and hard fats may be advantageously manufactured from saturated fats and fatty acids having about 10 to 18 carbon atoms per molecule. The method of the present invention contemplates reacting the aforementioned acids with a considerable excess of glycerol whereby the resultant esterified product has a hydroxyl value of between about 40 and 100, and preferably between 60 and 80.

[page 4]

Accordingly, a primary object of the present invention is to teach a method for the production of brittle and hard fats and the products obtained thereby.

It is another object of the present invention to teach the production of brittle and hard fats from saturated fatty acids by employing an excess amount of a polyhydric alcohol.

Additional objects and advantages of the present invention will become apparent from a detailed consideration of the following description.

The saturated acids that may be employed are preferably selected from the group comprising lauric acid, myristic acid, palmitic acid and stearic acid. These acids may be either employed alone or in admixture.

A unique feature of the present invention is in the fact that the brittle and hard fat produced does not completely have its melting point determined by the fatty acid or fatty acid mixtures employed. By utilizing an excess of glycerol one can considerably control and select a resultant product having a desired melting point.

If, for example, lower fatty acids, except lauric acid, derived from vegetable seed fat is hydrogenated to eliminate the unsaturated fatty acids, a fatty acid mixture is produced which has a melting point of about 40°C. Following this, if such a fatty acid mixture is esterified with such an excess of glycerol that the ester produced has a bydroxyl number of about 70, the melting point of the ester would then be between about 35 and 36°C. It will be appreciated that similar results may be obtained when other fatty acids mixtures are employed or technically pure lauric acid.

[page 5]

The melting point of the esterified product is dependent upon the amount of excess glycerin employed. For example, in the following table a comparison of hydroxyl numbers and melting points is made when the same amount of fatty acid, such as technically pure lauric acid, is esterified with glycerol.

TABLE

HYDROXYL NUMBER	MELTING POINT
6.2	40.3°C.
16.9	38.8
28.8	36.8
48.4	35.2
77.0	35.5
104.0	36.9
175.9	38.5
228.8	41.7

The above table shows clearly the meaning of the amount of glycerin in excess whereby a fatty product is produced having a desired melting point.

The inventive concept of the present invention permits the use of known steps to show an influence on the melting point of the end products by means of a clear manner of operation by which the quality of the end product may be easily predicted and varied.

The invention is illustrated but not limited by the following examples.

EXAMPLE I

Refined palm kernel fat is split and the fatty acids having fewer than 12 carbon atoms per molecule are removed. The fatty acid mixture is hydrogenated and distilled to produce a saturated fatty acid mixture having an acid number of 247.5, an iodine number of 1.9 and a melting point of 37.0°C. 180 kg. of the resultant saturated fatty acid mixture is reacted with 30 kg. of glycerol at a temperature of 210°C. under vacuum conditions in the presence of an esterification catalyst. The esterification is complete as soon as the acid number has a value below 0.5. The resultant fatty ester has a saponification index of 228, an iodine number of 2.1, a hydroxyl number of 50, and a melting point of 33.7°C.

EXAMPLE II

180 kg. of refined technically pure lauric acid having an acid number of 280, an iodine number of less than 1, and a melting point of 40.4°C. is esterified with 35 kg. glycerol in the manner described in connection with Example I. The resultant product has a saponification index of 256, an iodine number of less than 1, a hydroxyl number of 48 and a melting point of 35.2°C.

EXAMPLE III

180 kg. of a suitably purified lauric-myristic acid mixture having an acid number of 264 and a melting point of 40.5°C. is esterified with

34 kg. of glycerol. The end product has a saponification number of 240, an iodine number of less than 1, a hydroxyl number 79 and a melting point of 34.5°C.

EXAMPLE IV

180 kg. of a lauric-palmitic acid mixture having an acid number of 253 and a melting point of 39.4°C. is esterified with 33 kg. of glycerol in the manner described in connection with Example I. The resultant product has a saponification index of 229, an iodine number of less than 1, a hydroxyl number of 70 and a melting point of 34.3°C.

EXAMPLE V

180 kg. of a sample cut of a lauric-myristic-palmitic acid mixture fraction, having an acid number of 244 and a melting point of 39.0°C., is esterified with 32 kg. of glycerol. The resultant product has a saponification index of 228, an iodine value of less than 1, a hydroxyl number of 73 and a melting point of 35°C.

EXAMPLE VI

180 kg. of a mixture of low melting point distilled soap stock fatty acids containing technical stearic acid having an acid number of 241 and a melting point of 39.3°C. is esterified in the above described manner. The resultant product has a saponification index of 225, an iodine number of 1.9, a hydroxyl number of 70 and a melting point of 34.2°C.

It will be apparent that many changes and modifications of the several features of the process described herein may be made without departing from the spirit and scope of the invention. It is, therefore, to be understood that the foregoing description is by way of illustration of the invention rather than limitation upon the invention.

[101]
Div. 63

PLAINTIFF'S #1
Paper No. 25

MAILED

APR 30 1963

Appeal No. 423-39

amm

U. S. PATENT OFFICE
BOARD OF APPEALS

IN THE UNITED STATES PATENT OFFICE

BEFORE THE BOARD OF APPEALS

Ex parte Wolfgang Baenitz

- - -

Application for Patent filed February 12, 1959, Serial No. 792,706. Production of Fats.

Dicke & Craig for appellant.

Before Magil and Rosa, Examiners-in-Chief, and Behrens, Acting Examiner-in-Chief.

Behrens, Acting Examiner-in-Chief.

This is an appeal from the decision of the Examiner's refusal to allow claims 25 to 30 which were substituted for finally rejected claims 13 to 18.

Claim 25 which is representative reads as follows:

25. A method for controlling the melting point of hard and brittle fats within a relatively narrow desired range during the manufacture thereof by esterification of a fatty acid selected from the group consisting of saturated fatty acids having about 10 to 18 carbon atoms per molecule and mixtures thereof with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40

and 100, comprising the step of controlling the melting point of the ester mixture produced by said esterification by selecting the quantity of said excess glycerol.

[102] The following references are relied upon:

Christensen	2,022,494	Nov. 26, 1935
Kuhrt	2,634,278	Apr. 7, 1953
Feuge et al.	2,745,749	May 15, 1956

Appellant's esterification process and the process of the references are adequately described in appellant's Appeal Brief (pages 1 and 2) and the Examiner's Answer (pages 2 and 3), respectively.

Claims 25 to 30 were rejected as "lacking invention" over the Christensen patent or the Kuhrt patent especially in view of the Feuge _et_ _al_. patent. We are convinced that this rejection should be sustained because the claimed process, at the least, would have been obvious to a person of merely ordinary skill in the subject art from a consideration of these references.

Appellant's process involves esterification of the fatty acid or a mixture of fatty acids with an "excess" of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100. This process is said to comprise "controlling" the melting point of the ester mixture produced by said esterification by selecting the quantity of said excess glycerol.

We see no error in the Examiner's analysis of the prior art. The Christensen patent, for instance, directly illustrates a process meeting the requirements of the appealed claims. Christensen prepares mixed esters by the esterification of stearic acid with glycerol, or "glycerine." He may prepare either distearin or monostearin or mixtures of monostearin and distearin. He specifies that such mixtures may be prepared by regulating the quantity of glycerol reacted with the fatty acids. He specifies, for example (page 2, second column, lines 40 to 48), that by doubling the quantity of stearic acid a
[103] product consisting "mostly" of distearin can be obtained and that

various mixtures of mono- and distearin can be prepared by varying the quantity of stearic acid used, i.e., by varying the ratio of glycerol to higher fatty acids. It is known that stearin, monostearin and distearin vary in melting points and hydroxyl numbers, or hydroxyl content, as the Examiner states. Christensen indicates that a certain quantity of stearic acid is used for reaction with a certain quantity of glycerol so that Christensen controls the nature of the mixed ester product by controlling the quantity of excess glycerine employed. Since Christensen is preparing a mixture of distearin and monostearin, he employs an excess of glycerol over that required to prepare stearin.

Appellant's brief, indeed, makes this statement with regard to the knowledge of the prior art:

> "With regard to the novelty and patentability of the present invention, the applicant does not deny the pre-existence of knowledge, in the prior art, that fats may be obtained by esterifying fatty acids with glycerols even to the extent of producing a fat having a hydroxyl number of approximately 90-distearin."

Appellant argues that prior art does not suggest the "basic underlying concept of the present invention — namely, that the melting point of the ester mixture can be controlled during the esterification of the fatty acids by selecting a specific quantity of excess glycerol." Taking Christensen as representative of the prior art, Christensen selects a particular glycerol-fatty acid mixture to produce a specific stearin mixture. Christiansen was aware, or should have been aware, that the melting points of stearin and distearin are different and that mixtures of the two stearins vary in melting points with the composi-
[104] tion of the mixture. Appellant's claims call for a range of hydroxyl numbers of 40 to 100. Distearin, appellant states, has a hydroxyl number of approximately 90. Monostearin would be expected to have a higher hydroxyl number and stearin, an ester in which each of the

hydroxy groups is esterified, would be expected to have a lower hydroxyl number. The range called for, therefore, has not been shown to critically distinguish the claimed process from even the illustrative examples of Christensen.

It is apparent that the Kuhrt patent recognizes that a partial ester product will be in dynamic equilibrium with the alcohol (or glycerol), and the fatty reactant and that a change in the relative proportion of components will change the character of the product. (See column 3, line 15 et seq.) Following this teaching would not be a patentable expedient.

The decision of the Examiner is affirmed.

AFFIRMED

[Plaintiff's Exhibit 2]

MG = Molecular Weight
VZ = Saponification No.
OHZ = Hydroxyl (or OH) No.
Smp. = Melting Point

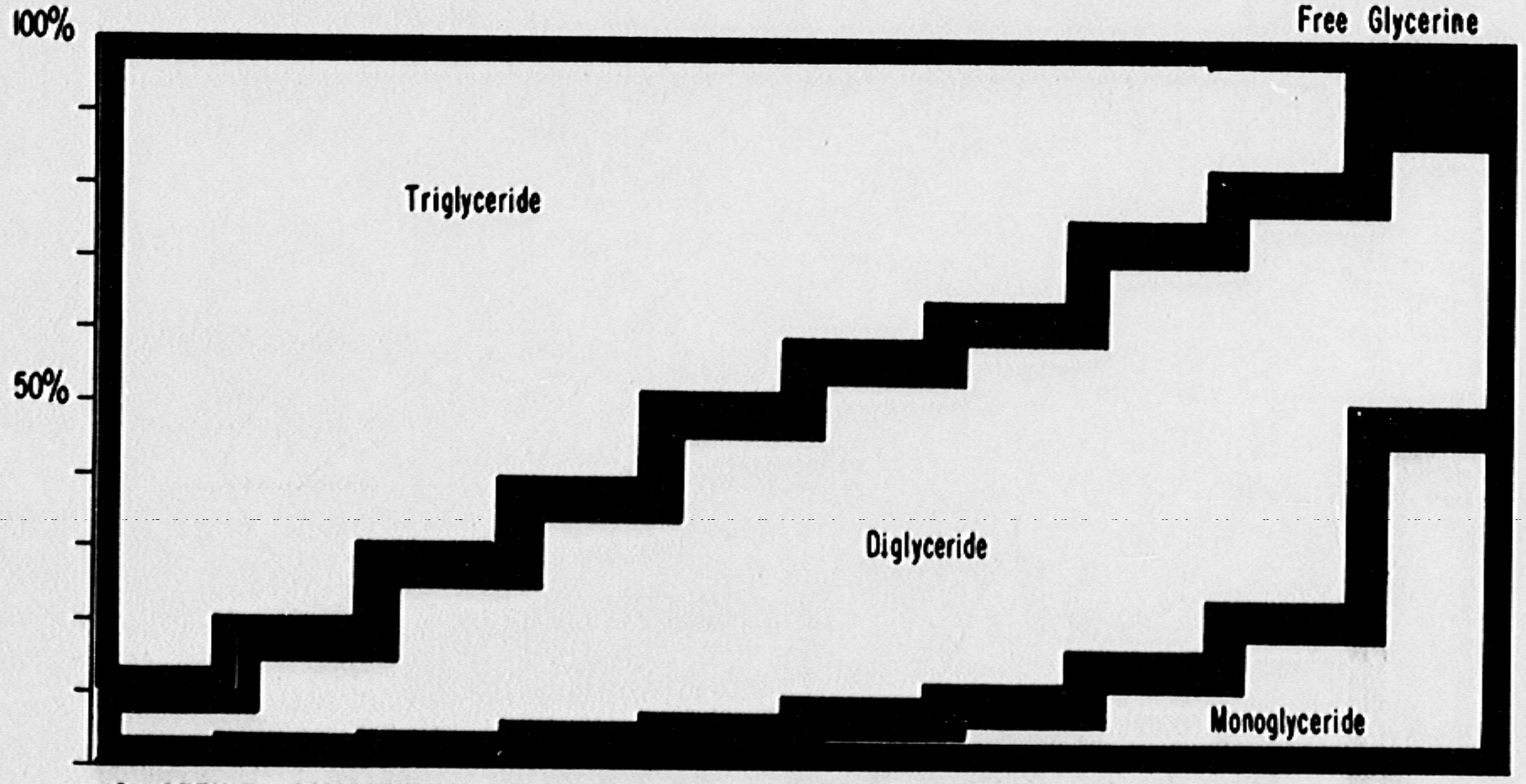
Fettsäure		Triglycerid				Diglycerid				Monoglycerid			
C-Zahl	MG	MG	VZ	OHZ	Smp.	MG	VZ	OHZ	Smp.	MG	VZ	OHZ	Smp.
10	172,26	554,82	303,3	0	31,5	400,58	280,1	140,1	44,5	246,34	227,7	455,4 ✓	53 ✓ 40,4
12	200,31	638,97	263,4	0	46,4 46,5	456,68	245,7	122,8	56,8 ~~39~~	274,39	204,9	409,0 ✓	63 ✓ 51,0;51,1
14	228,37	723,15	231,7	0	57,0	512,80	218,8	109,4	65,5 66,5 ✓ 59,5 59,4	302,45	185,4	371,1 ✓	70,5 ✓ 61,0; 61,2
16	256,42	807,30	208,6	0	65,5 66,4	568,90	197,2	98,6	72,5 ✓ 72,9 64 63,5	330,50	169,8	339,6 ✓	77 ✓ 68,5; 69
18	284,47	891,45	188,8	0	72,0 72,5 73,1	625,00	179,5	89,8	78 78,2 68,5 69 71	✓358,55	156,5	313,0 ✓	81,5 ✓ 79,4; 79,5

Erklärung: unter Diglyceride: die erstgenannten Schmelzpunkte gelten für die 1-3-Diglyceride, die dann aufgeführten für die 1,2-Diglyceride.

unter Monoglyceride: Die erstgenannten Schmelzpunkte gelten für 1-Monoglyceride, die dann aufgeführten für 2-Monoglyceride.

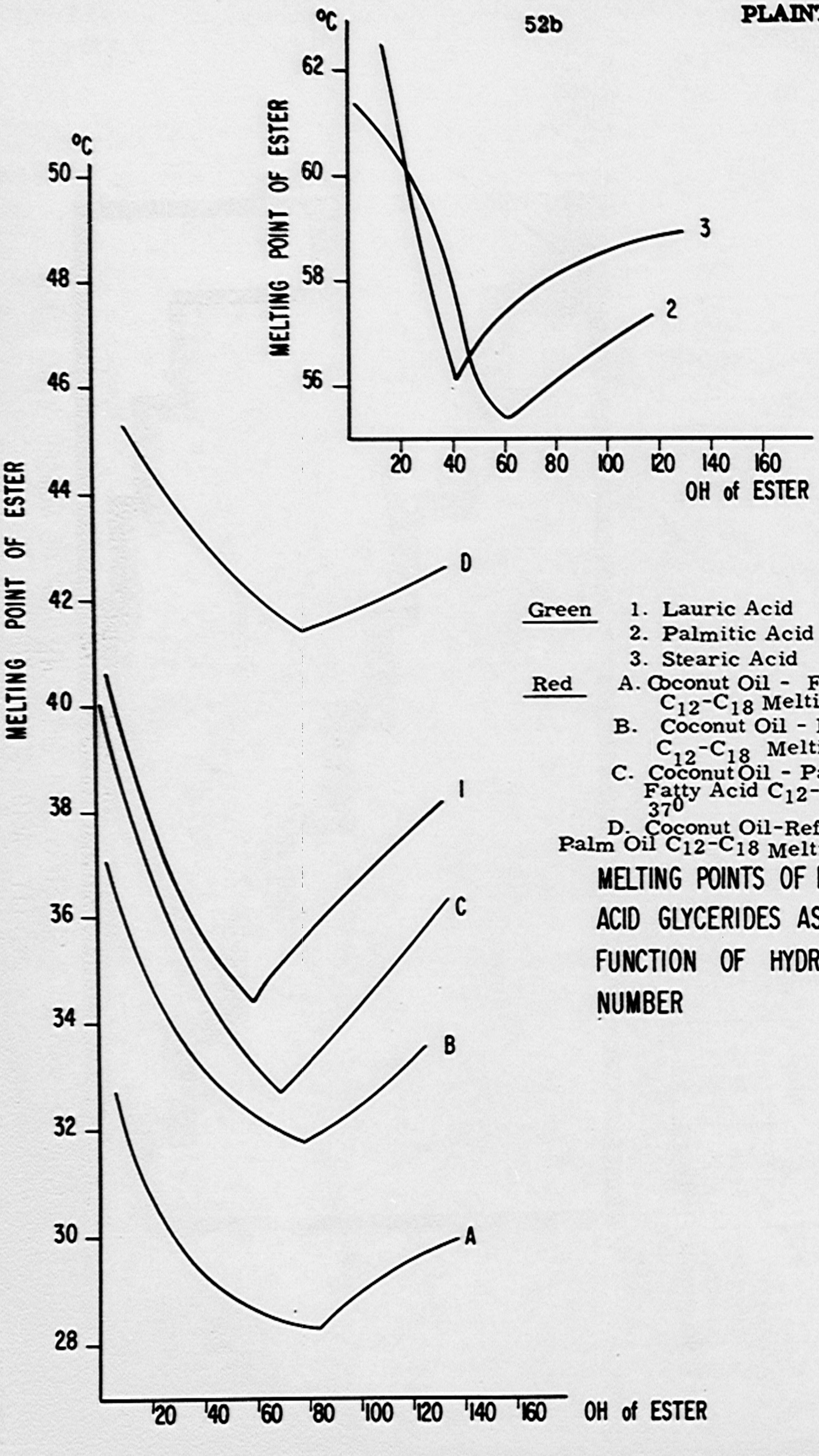
PLAINTIFF'S EXHIBIT #3

SZ	0.16	1.08	1.25	0.58	0.66	0.42	0.45	0.24	0.40	0.51
VZ	244.8	245.0	242.5	239.0	237.5	235.0	233.5	229.0	224.0	193.0
OHZ	12.4	22.2	31.8	49.5	68.4	82.6	93.2	120.0	149.0	351.0
Smp	32.4°	30.4°	29.6°	28.9°	28.4°	28.3°	28.9°	29.6°	31.0°	40.4°
% Free Fat	0.061	0.413	0.478	0.222	0.252	0.161	0.172	0.092	0.153	0.195
% Free Glycerine	0.0037	0.0065	0.014	0.020	0.032	0.018	0.027	0.045	0.11	6.15
% Triglyceride	89.58	82.53	72.93	63.74	51.86	44.51	39.12	27.97	20.96	5.45
% Diglyceride	10.25	16.18	24.80	33.30	43.30	48.60	52.40	58.70	58.40	41.81
% Monoglyceride	0.11	0.87	1.78	2.72	4.58	6.71	8.25	13.19	20.38	46.40



GLYCERINE COMPOSITION OF ESTERS WITH DIFFERENT HYDROXIL NUMBER WITH SAME FATTY ACID COMPOSITIONS

PLAINTIFF'S EXHIBIT #4

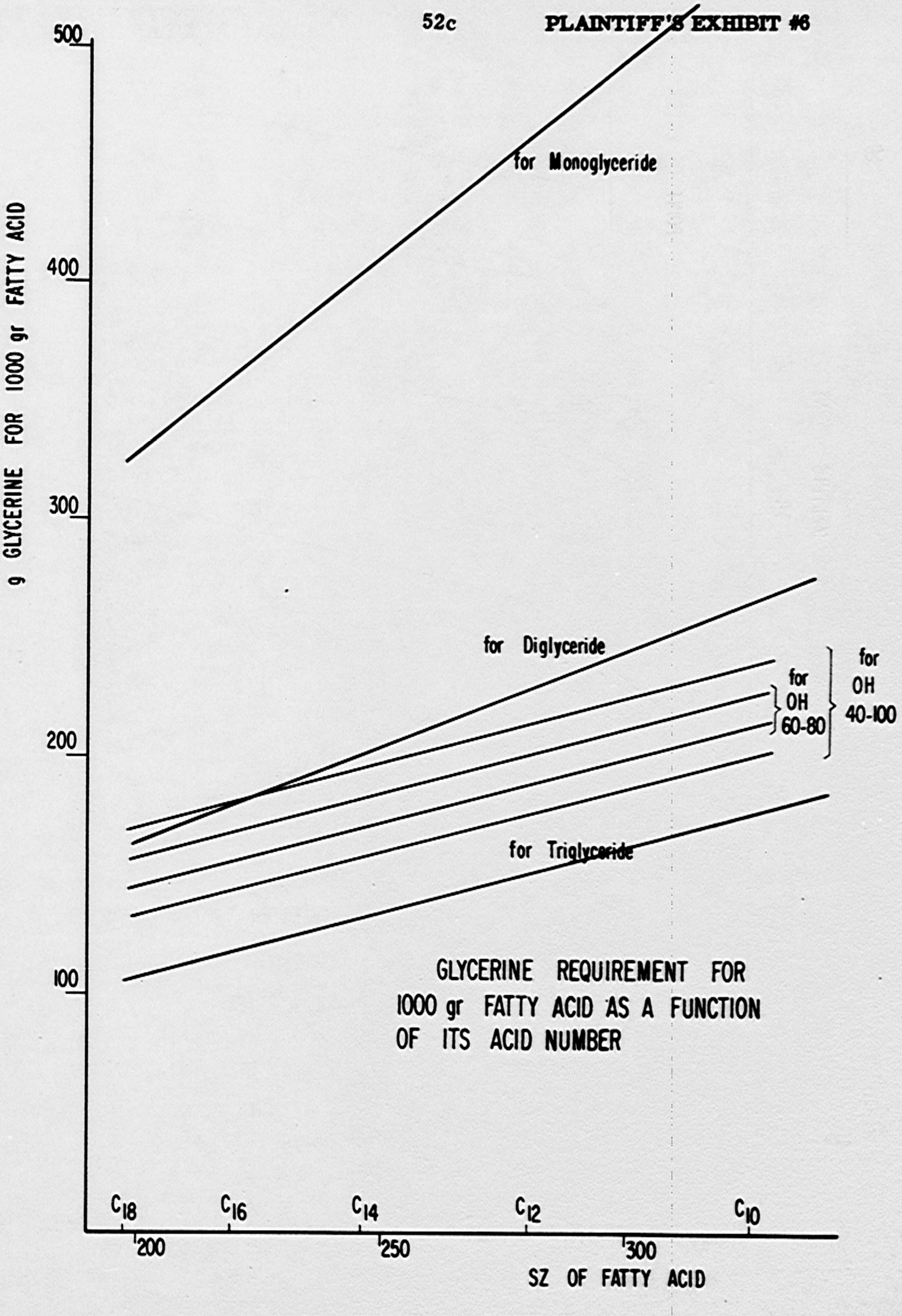


Green
1. Lauric Acid
2. Palmitic Acid
3. Stearic Acid

Red
A. Coconut Oil - Fatty Acid C_{12}-C_{18} Melting Point 28°-29°
B. Coconut Oil - Refined Fatty Acid C_{12}-C_{18} Melting Point 35.5°
C. Coconut Oil - Palm Kernel Refined Fatty Acid C_{12}-C_{18} Melting Point 37°
D. Coconut Oil-Refined Fatty Acid with Palm Oil C_{12}-C_{18} Melting Point 45.5°

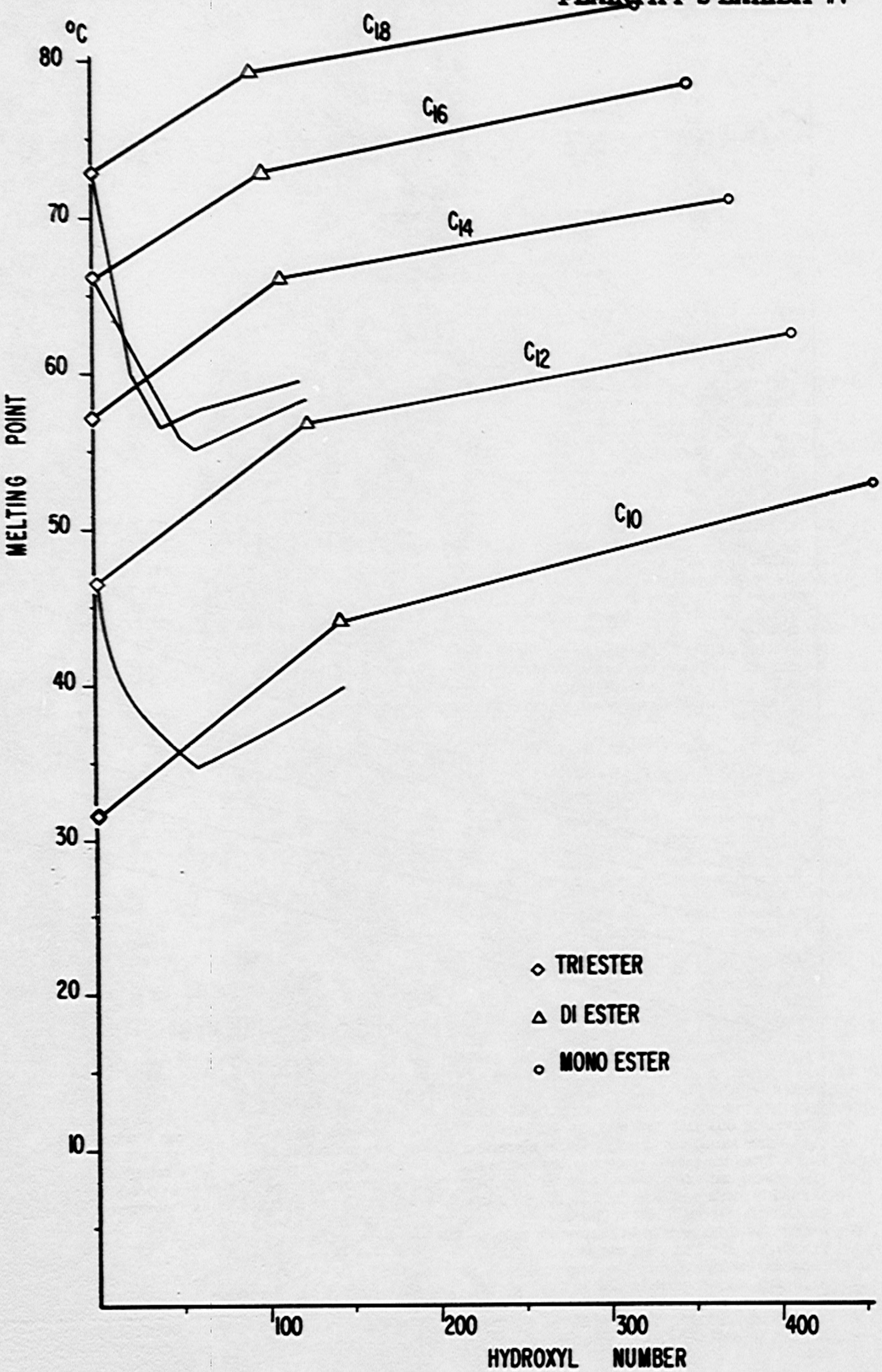
MELTING POINTS OF FATTY ACID GLYCERIDES AS A FUNCTION OF HYDROXIL NUMBER

PLAINTIFF'S EXHIBIT #6



GLYCERINE REQUIREMENT FOR 1000 gr FATTY ACID AS A FUNCTION OF ITS ACID NUMBER

PLAINTIFF'S EXHIBIT #7



[Defendant's Exhibit 1-A]

Patented Nov. 26, 1935 **2,022,494**

UNITED STATES PATENT OFFICE

2,022,494

PROCESS OF PREPARING MONO AND DI-GLYCERIDES

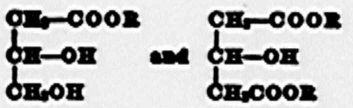
Carl W. Christensen, Chicago, Ill., assignor to Armour and Company, Chicago, Ill., a corporation of Illinois

No Drawing. Application February 14, 1935, Serial No. 6,501

12 Claims. (Cl. 87—12)

This invention relates to processes of preparing mono and diglycerides and it comprises processes wherein higher fatty acids are esterified in the presence of a catalyst of the group consisting of alkali-forming metal soaps of higher fatty acids and alkali-forming metal glycerates; it further comprises processes of incorporating mono and diglycerides in various fatty or oily materials such as vegetable oils, edible shortening materials, mineral oils and the like, wherein a higher fatty acid is esterified with glycerine in the presence of such fatty or oily material and in the presence of the above stated catalysts so that the resulting glyceride is formed in situ in the fatty or oily material.

Mono and diglycerides of higher fatty acids are becoming of increasing technical and practical interest because of their pronounced emulsifying power. These materials all possess the following type formula:



$$\begin{array}{l} CH_2\text{—}COOR \\ | \\ CH\text{—}OH \\ | \\ CH_2OH \end{array} \quad \text{and} \quad \begin{array}{l} CH_2\text{—}COOR \\ | \\ CH\text{—}OH \\ | \\ CH_2COOR \end{array}$$

monoglyceride diglyceride

wherein R is a relatively long hydrocarbon and COOR is a higher fatty acid radical. Whereas an ordinary vegetable or animal fat is a triglyceride, that is, there are no free hydroxyl groups in the glycerine molecule, the mono and diglycerides always contain at least one such free hydroxyl group and it is likely that their marked emulsifying power is due, at least in part, to this molecular structure.

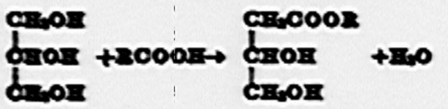
Within recent years it has been suggested to add a small amount of the mono and diglycerides to many different materials. For instance, the use of these glycerides in mineral oils has been patented. In this relation they confer beneficial properties on the resulting oil compound. For some time it has been known that edible oils such as shortening oils and fats can be improved by incorporating small amounts of these glycerides therein. Thus the melting point of the edible fat can be raised, and it can be made to emulsify more readily with water as in the manufacture of margarine. In still other instances, use of these particular glycerides in cosmetics, polishes, oily emulsions etc. has been suggested.

Curiously enough, while the triglycerides are the common major constituents in most all animal and vegetable fats, the mono and diglycerides do not exist in nature. Because of the fact that until quite recently they were largely laboratory curiosities, relatively few methods for preparing them have been described. The classical method is that of reacting a glyceryl chlorhydrin with a fatty acid; another is that of reacting glycerine with definite predetermined quantities of free fatty acid, there being just enough present to react with one or two of the hydroxyls of the glycerine. Neither of these methods have been satisfactory. They are difficult to control, relatively high temperatures are required, and yield has left much to be desired. Of late it has been discovered that glycerine can be made to react with a triglyceride whereby mono and diglycerides can be formed. This reaction, when catalyzed by certain catalysts, is of practical, commercial importance.

Although, as stated, attempts have been made to react glycerine with definite predetermined quantities of free fatty acid, the amount of acid being regulated to give either mono or diglycerides, this method has not been satisfactory because the reaction must be catalyzed. Hitherto the catalysts selected have invariably been acid reacting substances. While hydrochloric acid is, of course, a well known esterification catalyst for most esterifying reactions, it has never been particularly satisfactory in making mono and diglycerides. Generally, these acid catalysts seem to work best in the esterification of monohydric alcohols. The prior art has also proposed the use of Twitchell's reagent for catalyzing the reaction between glycerine and fatty acids, but here again no satisfactory commercial results have been obtained.

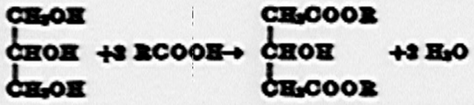
I have now discovered a method of preparing the mono and diglycerides which does use as starting substances glycerine and higher fatty acids. I have discovered that by changing the type of catalyst hitherto used I can obtain markedly superior results and avoid contamination of reaction products by undesirable by-products. This feature is of extreme importance when making mono and diglycerides because the major use for these materials is in the manufacture of edible products. Any process that requires considerable purification of the reaction product is not commercially feasible. I have discovered that the alkali-forming metal glycerates and the alkali-forming metal soaps are extremely good catalysts for the reaction between glycerine and higher fatty acids. I wish to distinguish at this time the process of the present invention from those processes which prepare mono and diglycerides by reacting glycerine with triglycerides. In the former instance, the reaction is a straight forward esterification of one or two of the hydroxyl groups of the glycerine. In the second case, the reaction takes an entirely different course. In it one or two of the higher fatty acid radicals of a triglyceride are removed therefrom. In a sense this is the reverse of esterification. In the present invention, as stated, I am directly reacting glycerine with a higher fatty acid and it is indeed surprising that alkali-reacting catalysts such as soaps and metal glycerates are good catalysts. It has hitherto always been considered that acid reacting catalysts must be used.

In its broadest aspects then my invention relates to reactions of the following type:



$$\begin{array}{l}CH_2OH\\ |\\ CHOH\\ |\\ CH_2OH\end{array} + RCOOH \rightarrow \begin{array}{l}CH_2COOR\\ |\\ CHOH\\ |\\ CH_2OH\end{array} + H_2O$$

leading to the formation of a monoglyceride and



$$\begin{array}{l}CH_2OH\\ |\\ CHOH\\ |\\ CH_2OH\end{array} + 2\,RCOOH \rightarrow \begin{array}{l}CH_2COOR\\ |\\ CHOH\\ |\\ CH_2COOR\end{array} + 2\,H_2O$$

giving diglycerides. In both instances RCOOH is a higher fatty acid. And in both reactions I use as catalysts alkali-forming metal glycerates or alkali-forming metal soaps or mixtures thereof.

Any of the higher fatty acids can be used in my process. By higher fatty acid I mean those fatty acids having six or more carbon atoms beginning with caproic. Thus, in the saturated series of fatty acids I can use caporic, caprylic, capric, lauric, myristic, palmitic, stearic, arachidic, behenic, carnaubic, cerotic, and melissic. These fatty acids are generally obtained from animal and vegetable sources in the well known ways. Caproic for instance is obtained from butter fat, others from cocoanut oil, stearic from tallow, and melissic from beeswax. I can use fatty acids of the oleic series of which oleic acid is the most important member, others being tiglic, physetoleic, rapic and erucic. The fatty acids of the linolic series are also suitable, the major one being linoleic and others being tariric and elaeomargaric.

As catalysts I can use any of the glycerates of the alkali or alkaline earth metals. Sodium glycerate has the greatest catalytic activity and it can be made in various ways. One of the easiest is to dissolve metallic sodium in glycerine to give either the mono or disodium derivative. Glycerine will also react with caustic soda, NaOH, to yield both the mono and di derivatives. The tri compound is unknown so far as I am aware, but there is no reason for supposing that it cannot be used. Potassium glycerates can be prepared in similar ways but the sodium compounds are considerably less expensive. Calcium glycerate can also be used and can be prepared in well known ways.

Of the soaps I find that sodium stearate is the cheapest and works the best although I can use other soaps of the alkali metals and also soaps such as calcium stearate. These soaps may be made from any of the higher fatty acids mentioned above and having six or more carbon atoms.

I shall now describe my invention with particular reference to the preparation of mono and distearin, it being understood, however, that I am not restricting myself thereto and that instead of starting with stearic acid I can start with any of the higher fatty acids noted above, or mixtures thereof.

For the preparation of monostearin I advantageously proceed as follows. I first dissolve with heating about two parts by weight of sodium hydroxide in 120 parts by weight of glycerine. In this way a small amount of sodium glycerate is formed in situ in a large volume of glycerine. This is a most convenient way of beginning but I can, of course, separately prepare substantially pure sodium glycerate and add it to my reaction mixtures. To the glycerine solution of sodium glycerate, heated to a temperature of about 150° C., I then add about three hundred parts by weight of triple pressed commercial stearic acid. During the addition of the stearic acid the mixture is advantageously stirred and I then increase the temperature to about 200° to 220° C. maintaining the heating for about an hour. At the end of this time the reaction mixture is permitted to cool. It sets to an amorphous wax-like solid and on analysis shows a free fatty acid content of about 3.4%. This indicates that the reaction has proceeded to such an extent that the yield is about 96–97% of the theoretical for monostearin.

In a similar manner I can prepare distearin by simply doubling the quantities of stearic acid used. Mixtures of mono and distearin can also be prepared by regulating the quantity of higher fatty acid reacted with the glycerine.

The above example refers specifically to the use of sodium glycerate as a catalyst. I shall now describe a further example wherein sodium stearate (soap) is used as a catalyst. In this example three hundred parts by weight of stearic acid are melted and then, with stirring, 120 parts by weight of glycerine containing 10 parts by weight of sodium stearate are added. The reaction mixture is heated to a temperature of about 200° to 220° C. over a period of an hour and then allowed to cool. Analysis of the reaction products shows that the yield of monostearin is about 93% of the theoretical. In a similar manner, by doubling the quantity of stearic acid a product consisting mostly of distearin can be obtained and various mixtures of mono and distearin can be prepared by varying the quantity of stearic acid used, in other words by varying the ratio of glycerine to higher fatty acids.

In the ordinary uses of the products of my process in the arts it is unnecessary to free them from any uncombined stearic acid and the catalysts used need not be removed. This is because the mono and diglycerides are generally incorporated in very small amounts in edible oils, etc., and the quantity of the catalyst, for example, is so small as to be quite harmless. However, the glycerides can be fractionally distilled from the soap catalyst under reduced pressure or they may be selectively dissolved from the soap. Ether, for example, will dissolve the glyceride but not the soap. Similar common laboratory expedients may be used for removing uncombined stearic acid from the reaction mixture.

It will, of course, be obvious that the process of the present invention is susceptible to many modifications. Thus, for instance, instead of using pure stearic acid I can use lard fatty acids, and I can use mixtures of stearic and palmitic, stearic and lauric and many others. In this way, I can obtain reaction products consisting of mixed mono and diglycerides.

My invention can be used as a method of incorporating mono and diglycerides in fats and oils in situ. This modification is particularly advantageous for margarine manufacture, in the preparation of shortening materials and the like.

For example, many edible oils contain quantities of free higher fatty acids. In such cases I simply add to such an oil or fat sufficient glycerine to convert the free fatty acid to a mono or diglyceride and also add a very small amount, of the order of 1% based on the glycerine, of an alkali-metal glycerate or a soap as a catalyst. The resulting mixture can then be heated to reaction temperature, of the order of 200° C., to cause the glycerine to esterify with the free fatty acids.

In another modification I can add higher fatty acid, glycerine, and catalyst to an edible oil or fat and heat the mixture to reaction temperature.

Both of these modifications just described give me finished oils or fats containing mono or diglycerides as desired, but generally I do not use them in my process because of the necessity for heating up a relatively large volume of fat or oil. The heating costs are inclined to be excessive. The process is of advantage, however, when it is desired to incorporate these glycerides in hydrogenated oils because the fatty acid, glycerine and catalyst can be added to the hot oil flowing from the hydrogenator, thus conserving the heat in the oil and putting it to good use. However, in those cases where it is rather difficult and expensive to free a fat or oil of higher fatty acids in the purification of the oil there are marked advantages in esterifying this higher fatty acid with glycerine in the presence of my catalysts.

Although I have specifically referred in the above examples to the use of soaps and glycerates of the alkali metals it will, of course, be understood that the alkaline earth metal compounds can also be used and accordingly in the appended claims I means the term glycerate or soap of an alkali-forming metal to generically designate both the alkali and alkaline earth groups. Likewise, "higher fatty acid" denotes those acids of the saturated and unsaturated series having six or more carbon atoms.

In the foregoing examples, I have specifically referred to a heating temperature of about 200–220° C. I do not wish to be limited to this particular temperature range, since the reaction between higher fatty acid and glycerine will proceed at a lower temperature, as low as 100–150° C., but at a considerably lower rate. Likewise, I can use temperatures somewhat in excess of 220° C., but ordinarily I do not find this to be particularly advantageous. At higher temperatures some slight decomposition, especially when using sodium glycerate as a catalyst, may occur. This is because of the tendency for lactic acid to be formed at relatively high temperatures.

Having thus described my invention what I claim is:

1. In the process of preparing fatty esters of the class consisting of mono and diglycerides by partially esterifying glycerine with higher fatty acids, the step which comprises partially esterifying the glycerine with a higher fatty acid at a temperature of about 100° C. to 220° C. in the presence of an alkali-forming metal glycerate as a catalyst.

2. In the process of preparing a monoglyceride, the steps which comprises esterifying glycerine with about the quantity of a higher fatty acid necessary to esterify one of the hydroxyl groups of the glycerine, the esterification being conducted at a temperature of about 100° C. to 220° C. in the presence of an alkali-forming metal glycerate as a catalyst.

3. The process as in claim 2 wherein the catalyst is sodium glycerate.

4. The process as in claim 2 wherein the fatty acid is lard fatty acids.

5. In the process of preparing a diglyceride, the steps which comprises esterifying glycerine with about the quantity of a higher fatty acid necessary to esterify two of the hydroxyl groups of the glycerine, the esterification being conducted at a temperature of about 100° C. to 220° C. in the presence of an alkali-forming metal glycerate as a catalyst.

6. The process as in claim 5 wherein the catalyst is sodium glycerate.

7. The process as in claim 5 wherein the fatty acid is lard fatty acids.

8. The process of preparing monostearin which comprises heating to a temperature of about 200–220° C. a mixture of glycerine, stearic acid and a catalyst comprising an alkali-forming metal glycerate, the stearic acid being in about the quantity necessary to esterify one of the hydroxyl groups of the glycerine.

9. The process as in claim 8 wherein the catalyst is sodium glycerate.

10. The process of preparing distearin which comprises heating to a temperature of about 200–220° C. a mixture of glycerine, stearic acid and a catalyst comprising an alkali-forming metal glycerate, the stearic acid being in about the quantity necessary to esterify two of the hydroxyl groups of the glycerine.

11. The process as in claim 10 wherein the catalyst is sodium glycerate.

12. The process as in claim 1 wherein the catalyst is sodium glycerate.

13. In the process of preparing fatty esters of the class consisting of mono and diglycerides by partially esterifying glycerine with higher fatty acids, the step which comprises heating a mixture containing glycerine, higher fatty acid, and a catalyst comprising an alkali-forming metal glycerate, at a temperature of about 100° C. to 220° C., the ratio of glycerine and fatty acid being such that the glycerine is but partially esterified to form the said fatty esters.

CARL W. CHRISTENSEN.

[Defendant's Exhibit 1-B]

United States Patent Office

2,745,749
Patented May 15, 1956

2,745,749

GLYCERIDIC MIXTURES EXHIBITING UNIQUE PROPERTIES AND PROCESS FOR THEIR PRODUCTION

Reuben O. Feuge, New Orleans, Earl J. Vicknair, Marrero, and Klare S. Markley, New Orleans, La., assignors to the United States of America as represented by the Secretary of Agriculture

No Drawing. Application July 19, 1951,
Serial No. 237,658

12 Claims. (Cl. 99—118)

(Granted under Title 35, U. S. Code (1952), sec. 266)

The invention herein described may be manufactured by or for the Government of the United States of America for governmental purposes throughout the world without the payment to us of any royalty thereon.

This invention relates to a process for chemically modifying certain glyceridic mixtures so that the modified mixture exhibits the properties of texture, melting point, flexibility and the like desired for a particular utilization. More particularly, the invention provides glyceridic mixtures exhibiting unique and valuable properties; and provides a process for their production by the controlled acylation of mixtures which are composed of glycerides of long-chain fatty acids and which contain substantial proportions of monoglycerides.

Glyceridic mixtures (i. e. fats) having the properties of texture, melting point, flexibility and the like suitable for a given use and also retaining the chemical properties of mixtures of saturated fatty acid esters of glycerol have a wide range of valuable application. Such mixtures are miscible with many organic compounds by virtue of their high carboalkoxy group content and are relatively resistant to oxidation by virtue of the fact that their hydrocarbon chains are saturated aliphatic chains. The mixtures provide particularly valuable plasticizers, lubricants and the like. Where the mixtures contain no acyl radicals exhibiting toxic physiological properties, they are edible fats and have unique value in food and drug applications.

The step of acylating a mixture of glycerides containing some monoglycerides is not in itself new. Some commonly used methods of determining the hydroxyl value of a fat or oil involve its acylation. However, in such processes, the acylation is conducted solely for the purpose of determining this analytical value of the glyceridic mixture. To accomplish this, a small amount of fat is reacted with a large excess of acylating agent and the product so produced is immediately dissolved or in other ways disposed of so that the amount of acylating agent used up in the reaction can be determined.

We have discovered that when a mixture of glycerides of long chain saturated fatty acids containing substantial portions of long chain monoglycerides is acylated under controlled conditions with the introduction of saturated fatty acid acyl groups of relatively short chain length, i. e., two to six carbon atoms, so that only a portion of the hydroxyl groups contained in the mixture are acylated while the mixture is unchanged in its chemical nature in that it is still a mixture of glycerides of saturated fatty acids, the mixture exhibits totally different properties of texture, melting point, flexibility and the like.

For example, when mixtures of glycerides principally consisting of monostearates or mixtures of monostearates and monopalmitates are subjected to acylation, under conditions not conducive to interesterification, until the hydroxyl value of the acylated mixture is between about 15 to 200 and the monoglyceride content of the acylated mixture is between about 0.1 to 30% a unique edible fat is produced. While in fats, properties of nongreasiness and flexibility are normally mutually exclusive, the edible fats so produced are either nongreasy, flexible solids or liquids.

Mixtures of glycerides, which contain at least 50% monoglycerides, in which stearates or stearates and palmitates constitute the only components present in more than small amounts, constitute the preferred starting materials for employment in the present process. As is well known to those skilled in the art, such mixtures vary in their physical properties. Where it is desired to impart to such mixtures (e. g., those containing relatively large amounts of the higher melting di- and triglycerides) a high degree of flexibility by the process of the present invention, it is preferable that the acylation be continued until the monoglyceride content is reduced to a relatively low value.

The step of acylating the glyceridic mixtures in accordance with the process of the present invention can be conducted in any conventional manner involving acylating agents and conditions which are not conducive to high rates of interesterification. Acids, acid anhydrides and acyl halides can suitably be employed as the acylating agents. The reaction conditions most conducive to acylation in the absence of interesterification are well known to those skilled in the art.

The choice of the individual acyl radicals introduced markedly affects the properties of melting point, flexibility and the like of the acylated mixture. The acyl radicals of the lower molecular weight anhydride-forming fatty acids, particularly those of acetic, propionic and butyric acid are the ones preferred for introduction in accordance with the process of the present invention. While acylated mixtures melting above normal room temperature can readily be produced by the introduction of the proper amounts of acetyl and propionyl radicals, the introduction of appreciable amounts of butyryl radicals tends to produce mixtures which are liquid at room temperature.

The acylation reaction involved in the process of the invention is controlled by the selection of reaction conditions favoring acylation over interesterification and by sampling and analyzing the acylated mixture for hydroxyl value and monoglyceride content. Sampling and analyses can be conducted in the conventional manner. Use of about the stoichiometric equivalent or less of the acylating agent is preferred. The reaction can be conducted in a continuous or batchwise manner under normal, super or subatmospheric pressure.

The following examples are presented to illustrate in more detail certain features involved in the practice of the invention. However, as it is apparent that numerous variations can be made, the scope of the invention is defined by the claims and is not to be construed as being limited to the particular materials and conditions recited in the examples.

Three mixtures composed of stearates and mixed stearates and palmitates were acetylated in accordance with the particularly preferred method of conducting the process of the invention to form a series of 9 acetylated mixtures having hydroxyl values of from about 35 to 170 and monoglyceride contents of from about 0.1 to 20%. These acetylated mixtures constitute particularly preferred flexible solid compositions provided by the invention. They are edible and nongreasy. They have many potential uses in the food industry, for example, as coatings for eviscerated poultry, other meat products, cheese, candies ice cream bars and the like. They are as easy to apply as paraffin and have the same texture and feel, but are at least six times more flexible. They are impervious to moisture and resistant to the attack of molds and bacteria.

The mixtures which were so acetylated are hereinafter referred to as monostearates A, B, and C. Monostearate A was prepared from pure stearic acid by the method of Gros et al. (J. Am. Oil Chemists' Soc., 28, 1–4, 1951); it contained 99.2% monostearate by analysis (Handschumaker, E., and Linteris, L., J. Am. Oil Chemists' Soc., 24, 143–145, 1947) and had a hydroxyl value of 306.5 according to the acetylation method of West et al. (J. Biol. Chem., 104, 627–634, 1934) modified by using one part of acetic anhydride to three parts of pyridine. Monostearate B was a commercial, molecularly distilled product. It had a hydroxyl value of 335.5 and a monoglyceride content of 91.5%. The average molecular weight of the combined fatty acids in this product was 270.5, which corresponds to a 1:1 ratio of stearic and palmitic acids. Monostearate C was a technical grade product made from completely hydrogenated cottonseed oil. Its hydroxyl value was 236.9, and it contained 61.0% monoglycerides of fatty acids having an average molecular weight of 280.0.

The monostearates were acetylated in the following manner to produce the acetylated mixtures hereinafter referred to as acetostearates, those from monostearate A being designated by A–1, A–2, those from monostearate B being designated by B–4, B–5, etc. The acetostearates were prepared by reacting acetic anhydride with monostearate. A temperature of 110° C. and a reaction time of one hour was used in most instances. The reactants were stirred and were kept under dry hydrogen to prevent reaction of the acetic anhydride with atmospheric moisture. The reaction was interrupted as desired by adding hot water to the reaction mixture and stirring for 10 minutes to hydrolyze the unreacted acetic anhydride. After washing the reaction product with distilled water it was dried by warming it under reduced pressure and stripping with hydrogen.

The physical properties of the acetostearates formed by reaction of monostearate with acetic anhydride could be controlled to a considerable extent by manipulating the reaction time, temperature, and proportion of acetic anhydride employed. None of the reactions was carried to completion, as evidenced by the hydroxyl value of the finished products.

The ratios of monostearate to acetic anhydride used in making the products subjected to elongation and bending tests were such that one equivalent of hydroxyl in the monostearate was mixed with 0.5, 1.0, and 2.0 moles of acetic anhydride, respectively. The reactions were stopped after one hour at 110° C.

The free fatty acid content of the acetostearates was nearly always between 1.0 and 1.5%, which is quite low in view of the fact that two of the monostearates used to prepare the products contained several tenths of a percent of free fatty acids. Therefore, it must be concluded that reaction occurred by direct acetylation, uncomplicated by interesterification. The short melting range of the products prepared with monostearates A and B, which in one case was less than a degree, and the fact that monostearates are relatively stable when heated to 110° C. in glacial acetic acid substantiate the validity of this conclusion.

The flexibilities of the acetostearates were measured by stretching test samples of each product. This was accomplished by heating the acetostearates to 60° C., pouring them into a mold at the same temperature, and solidifying them by cooling to room temperature (26° C.). After being removed from the mold, the samples were held overnight at 22° C. and tested at the same temperature.

The test samples were formed by casting the melted product in a mold consisting of three plates of ⅛-inch sheet aluminum separated by aluminum foil. The center plate, or mold proper, was cut out to form a ribbon shaped opening. The other two plates were confining faces of the mold.

Each molded test sample measured five inches in length and ⅛ inch in thickness. The center portion of the sample was ¾ inch in width, but one inch from each end the width increased gradually so that the ends were 1¼ inches wide. In making the tests the enlarged end-sections of the sample were covered with fine sand paper and clamped in the jaws of the testing machine. The central section of the sample measuring 3 x ¾ x ⅛ inch was subjected to stretching.

An Instron Tensile Tester was used to stretch the samples. In this machine one of the jaws is pulled away from the other at a contant rate, while the machine automatically records the load or pull necessary to maintain the rate of elongation and synchronizes this value with a recording of the amount of elongation or stretch of the test sample.

The results of the tests made by applying a stretching rate of 1.0 inch per minute are recorded in Table I. Data for tests made with monostearate B and a paraffin (M. P., 50–52° C.) are also included in the table.

TABLE I

Elongation at break point and resistance to stretching of acetostearates, monostearate, and paraffin

Product No.	Mole $(CH_3CO)_2O$/ OH equivalent *	Maximum load, lbs.	Load at break, lbs.	Elongation, Percent
A–1	0.5	1.7	----------	>800
A–2	1.0	0.71	----------	>800
A–3	2.0	0.40	----------	>800
B–4	0.5	1.6	0	467
B–5	1.0	0.43	0	313
B–6	2.0	0.22	----------	>800
C–7	0.5	8.8	8.1	31
C–8	1.0	6.7	0.3	261
C–9	2.0	1.6	----------	>800
Monostearate B	----------	2.7	2.7	4
Paraffin	----------	8.0	8.0	5

* Ratios are moles of acetic anhydride used per OH equivalent in the preparation of the acetostearates.

Each acetostearate shown in Table I stretches far more than either monostearate B or the paraffin. More than half of the acetostearates stretched over 800% which was the limit of the testing machine. The lowest value observed for any of the acetostearates was six times greater than was obtained with paraffin. From the data for the acetostearates prepared from monostearates B and C, it is apparent that the stretchability increased as the proportion of acetic anhydride used to prepare a given product increased.

The relative tenacity or resistance to stretching, which is indicated by the maximum load values in Table I, varied greatly, depending on the type of monostearate and proportion of acetic anhydride employed in the preparation of the acetostearates. The higher proportions of acetic anhydride gave products with the least tenacity because they acetylated greater proportions of the monostearates and transformed them into lower melting compounds.

Products made with monostearate B were not as tenacious as the corresponding products made with monostearate A, probably because the former contained monoglycerides of palmitic and possibly other fatty acids. The use of monostearate C resulted in the toughest products because it contained a large proportion of diglycerides.

The maximum loads shown in Table I are numerically equal to about 1/11 of the stress in pounds per square inch which must be applied to the molded product to start stretching at a rate of 33.3% per minute. A force of 73.7 pounds per square inch had to be applied, for example, to product No. 8 to start it stretching at a rate of 33.3% per minute.

Several of the acetostearates were subjected to stretching at rates other than one inch per minute. The results of these tests are shown in Table II. From the data in Table II it can be seen that even at greatly increased rates of elongation the products flowed like liquids. A 20-fold increase in the rate of elongation resulted in about a 3-fold increase in the load or force necessary to overcome the resistance to stretching. Increased rates of elongation decreased the amount of elongation before the sample ruptured.

TABLE II

Elongation at break point and resistance to stretching of acetostearates under different rates of elongation

Product No.	Rate of elongation, in./min.	Maximum load, lbs.	Load at break, lbs.	Elongation, Percent
A-1	1	1.7	--------	>800
	5	3.2	0	>800
	20	4.4	0	547
A-2	1	0.71	--------	>800
	20	2.2	0	630
A-3	1	0.40	--------	>800
	20	1.6	--------	>800
B-4	1	1.6	0	467
	10	3.2	0	350
C-9	1	1.6	--------	>800
	5	3.0	0.3	613
	10	4.2	0.8	380
	20	5.4	2.2	173

In order to obtain date on the flexibility on the acetostearates at a temperature other than that of the testing laboratory (22° C.) in which the Instron Tensile Tester was located, bending tests were made at 4° C. on several of the products listed in Table I. For these tests a 5 x ¾ x ⅛ inch ribbon of acetostearate was molded on a strip of high-strength filter paper by using a molding technique similar to that described, except that a sheet of filter paper was substituted for one of the sheets of aluminum foil. The molded product was maintained at 4° C. overnight before testing.

The bending test consisted of fastening one end of the ribbon of acetostearate and filter paper to a brass cylinder one inch in diameter and rotating the cylinder at a rate of 33° per minute while pulling at the other end of the ribbon with a force of two pounds.

Paraffin and monostearate B cracked when tested in this manner. Acetostearates Nos. C-7 and C-8 also cracked; but product Nos. A-2, A-3, B-4, B-5, and B-6 did not crack. Acetostearates Nos. A-1 and C-9 cracked only slightly.

The acetostearates made with monostearate B were also tested in another way, which consisted of placing strips of the products in the freezing compartment of a domestic refrigerator. The products, especially B-5 and B-6 remained pliable at the freezing temperatures.

The melting range of a coating fat is an important physical property. This property of the acetostearates is about as important as flexibility, especially if the products are to be used as edible coatings.

The nine acetostearates listed in Table I were nongreasy solids at room temperature (26° C.), but their melting ranges, determined by the capillary tube method, differed appreciably. The melting ranges, hydroxyl values, and monoglyceride contents of the products are recorded in Table III.

TABLE III

Melting range, hydroxyl value and monoglyceride content of acetostearates

Product No.	Melting range, ° C.	Hydroxyl value	Monoglyceride content, percent
A-1	45.0–48.0	153	17.8
A-2	39.5–42.0	78	2.7
A-3	38.0–39.0	36	0.3
B-4	41.0–44.0	105	11.4
B-5	32.0–35.5	78	2.8
B-6	29.0–31.0	36	0.9
C-7	46.0–54.0	122	14.1
C-8	28.0–54.0	84	4.9
C-9	25.0–54.0	38	0.4

The lower temperature recorded for each product is the temperature at which melting was first observed; the higher temperature is the one at which the product was completely liquid.

Product No. B-6 began melting at 29° C. and product No. A-1 began melting at 45° C. All of the products except those made with monostearate C have relatively short melting ranges. The shortest melting range observed (product No. A-3) was less than one degree.

While the above glyceridic mixtures having hydroxyl values above about 35 are solids at normal room temperature, glyceridic mixtures having hydroxyl values below about 35, which mixtures are liquid at normal room temperatures, are also provided by the invention. For example, monostearate B was acetylated by the process described above to a hydroxyl value of about 20 and a monoglyceride content of about 0.1. The mixture was a unique completely saturated, colorless, odorless and tasteless oil which exhibited an exceptionally high resistance toward degradation induced by heat and oxidative conditions. When enough propyl gallate was added to this mixture to prevent the oxidation of the oleoyl radical (present due to the fact that the monostearate used contained about 3% olein) the mixture, when subjected to aeration for more than 700 hours at 208° F., exhibited a peroxide value of less than about 3 milli-equivalents of peroxide per kiligram of fat (Accelerated Stability Test Using Peroxide Value as an Index, A. E. King et al. J. Am. Oil Chem. Soc. 10 105–9, 1933) and exhibited no change in color, odor or taste.

Having thus described our invention, we claim:

1. A process which comprises acylating a mixture containing at least 50% monoglycerides of the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof with an acylating agent of an unsubstituted saturated alkanoic acid containing from 2 to 4 carbon atoms, and continuing the acylation until a product is obtained having a hydroxyl value of from 15 to 200 and a residual monoglyceride content of from 0.1 to 30%.

2. A process for the production of an edible flexible solid composition which comprises acetylating, a mixture containing at least 50% monoglycerides of the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof until the acetylated mixture has a hydroxyl value of from about 37 to 170 and a residual monoglyceride content of from about 0.1 to 20%.

3. A mixture of glycerides comprising from about 0.1 to 30% of monoglycerides from the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof and glycerides in which a portion of the hydroxyl groups are acylated by an unsubstituted saturated fatty acid radical of not more than 4 carbon atoms, said mixture having a hydroxyl value of from 15 to 200.

4. A flexible solid composition comprising a mixture of glycerides containing from about 0.1 to 30% of monoglycerides from the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixture thereof and glycerides in which a portion of the hydroxyl groups is acylated by acetyl radicals, said mixture having a hydroxyl value of from 35 to 170.

5. A liquid mixture of glycerides comprising from about 0.1 to 30% monoglycerides from the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof and glycerides in which a portion of the hydroxyl groups is acylated by acetyl radicals, said liquid mixture having a hydroxyl value of from about 15 to 35.

6. A heat-resistant and oxidation-resistant liquid fat comprising a mixture of glycerides containing about 0.1% of monoglycerides of the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof and glycerides in which a portion of the hydroxyl groups is acylated by acetyl radicals, said mixture having a hydroxyl value of about 20.

7. A process which comprises acylating a mixture containing at least 50% of monoglycerides of the group con-

sisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof with an acylating agent of an unsubstituted saturated alkanoic acid containing from 2 to 4 carbon atoms, continuing the acylation until a product is obtained having a hydroxyl value of from 15 to 200 and a residual monoglyceride content of from 0.1 to 30%, and coating an edible article with the acylated product.

8. A process which comprises reacting a fatty mixture containing at least 50% monoglycerides from the group consisting of glyceryl monostearate, glyceryl monopalmitate and mixtures thereof with acetic anhydride, continuing the reaction until the product has a hydroxyl value of from 15 to 200 and a residual monoglyceride content of from 0.1 to 30%, stopping the reaction by adding hot water when the product has attained the aforementioned hydroxyl value and monoglyceride content, and recovering said product from the reaction mixture.

9. The process of claim 1 in which the acylating agent is acetic anhydride.

10. The process of claim 1 in which the acylating agent is an acetylating agent.

11. The process of claim 3 in which the acetylating agent is acetic anhydride.

12. A process of protecting an edible article, which process comprises, coating the article with the composition of claim 5.

References Cited in the file of this patent

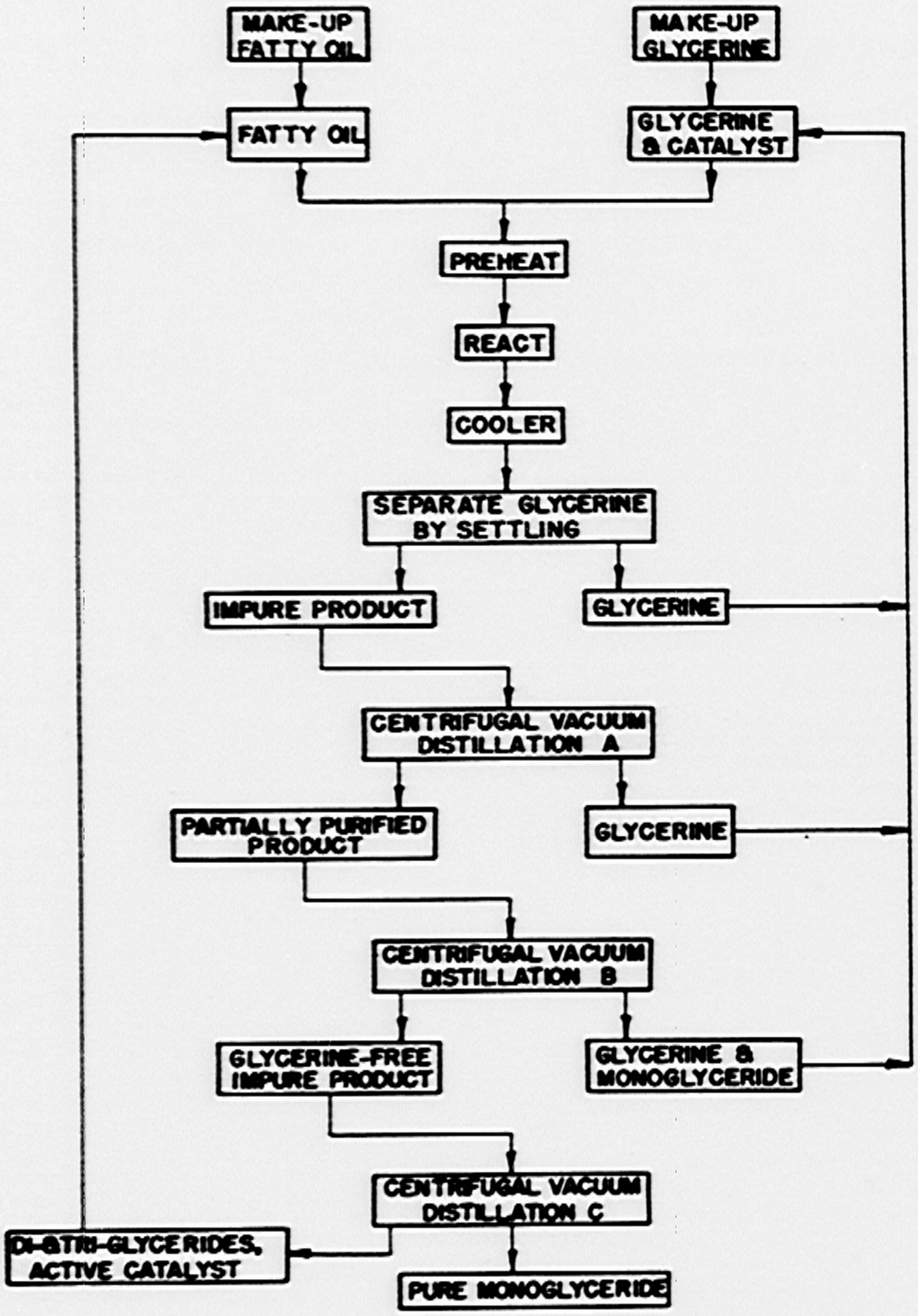
UNITED STATES PATENTS

2,145,443	Harris	Jan. 31, 1939
2,160,532	Barrett	May 30, 1939
2,322,198	Parsons	June 15, 1943
2,584,998	Filachione et al.	Feb. 12, 1952
2,615,159	Jackson	Oct. 21, 1952
2,615,160	Baur	Oct. 21, 1952
2,690,971	Iveson	Oct. 5, 1954

[Defendant's Exhibit 1-C]

April 7, 1953 N. H. KUHRT 2,634,278

PROCESS FOR SUBSTANTIALLY COMPLETE CONVERSION
OF FATTY MATERIAL TO PARTIAL ESTER
Filed July 16, 1949


MAKE-UP FATTY OIL
MAKE-UP GLYCERINE
FATTY OIL
GLYCERINE & CATALYST
PREHEAT
REACT
COOLER
SEPARATE GLYCERINE BY SETTLING
IMPURE PRODUCT
GLYCERINE
CENTRIFUGAL VACUUM DISTILLATION A
PARTIALLY PURIFIED PRODUCT
GLYCERINE
CENTRIFUGAL VACUUM DISTILLATION B
GLYCERINE-FREE IMPURE PRODUCT
GLYCERINE & MONOGLYCERIDE
CENTRIFUGAL VACUUM DISTILLATION C
DI-&TRI-GLYCERIDES, ACTIVE CATALYST
PURE MONOGLYCERIDE

INVENTOR
NOEL H. KUHRT
BY
George W. Petersen
ATTORNEYS

Patented Apr. 7, 1953 2,634,278

UNITED STATES PATENT OFFICE

2,634,278

PROCESS FOR SUBSTANTIALLY COMPLETE CONVERSION OF FATTY MATERIAL TO PARTIAL ESTER

Noel H. Kuhrt, Rochester, N. Y., assignor, by mesne assignments, to Eastman Kodak Company, Rochester, N. Y., a corporation of New Jersey

Application July 16, 1949, Serial No. 105,182

19 Claims. (Cl. 260—410.7)

This invention relates to a process of treating fatty material and is particularly concerned with a process of effecting conversion of fatty material to a fatty acid partial ester of a polyhydric alcohol.

Fatty acid partial esters of polyhydric alcohols are prepared by reacting fatty material with an alcohol in the presence of an ester-interchange catalyst.

It is an object of this invention to provide a highly effective process for treating fatty material.

It is a further object of the invention to provide a commercially feasible process for preparing fatty acid partial esters of polyhydric alcohols.

Another object of the invention is to provide a process for effecting substantially complete conversion of fatty material to a partial ester.

Another object is to provide a novel process whereby substantially pure partial ester is prepared and recovered.

A further object of the invention is to provide a process whereby ester-interchange catalyst is retained in active form throughout the process.

Another object of the invention is to provide a cyclic process of preparing monoesters.

Another object of the invention is to obtain monoglycerides in high yield.

Another object of the invention is to provide a process whereby substantially all of the partial ester formed by alcoholysis of fatty material is recovered.

Other objects will be apparent from the drawing, description and claims.

These and other objects are attained by means of this invention which is best illustrated by reference to an embodiment thereof. The single figure of the drawings is a schematic representation of a process embodying the invention whereby a fatty oil such as soybean oil is converted to monoglyceride by reacting the soybean oil with glycerine in the presence of an ester-interchange catalyst.

In converting soybean oil to monoglyceride in accordance with this invention, a predetermined amount of the oil is admixed with an excess of glycerine and with an ester-interchange catalyst. The amount of glycerine employed for alcoholysis of the oil may be varied; and, although stoichiometric or smaller amounts may be employed, it is preferable to use an excess of glycerine for optimum conversion of the oil to monoglyceride. The convertible fatty material in the soybean oil consists largely of fatty acid compounds present as free fatty acid or fatty acid esters, particularly

triglycerides, and the amount of glycerine used is calculated from the amount of convertible fatty material present in the oil calculated as fatty acid. Thus the glycerine may be used in stoichiometric amounts or less but is preferably employed in at least 40% excess over stoichiometric amounts and in many cases as much as 200% excess or more.

The alcoholysis is carried out in the presence of an ester-interchange catalyst which promotes the alcoholysis or esterification. Suitable ester-interchange catalysts include any of the well-known alkaline or acid catalysts which do not distill simultaneously with the desired partial ester. Catalysts desirably employed in the alcoholysis of soybean oil or similar vegetable oil or other fatty material are the metal compounds which react with fatty acid present in the reaction mixture to form a metal soap which functions as ester-interchange catalyst. Particularly suitable metal compounds are the alkali and alkaline earth metal oxides and hydroxides and desirably the bivalent metal compounds. Thus although such monovalent metal compounds as sodium hydroxide, sodium methylate, sodium carbonate and the like may be employed, particularly efficacious results are obtained by adding to the reaction mixture such bivalent metal compounds as strontium oxide, calcium oxide, calcium hydroxide, strontium hydroxide, barium oxide and the like. Strontium compounds are particularly desirable catalytic materials because of the enhanced yield of partial ester and its marked ability to prevent formation of color bodies during the process. The absence of color body formation is of particular importance in the cyclic process and particularly a continuously circulating process since color bodies formed in the process otherwise build up in the system and contaminate the product.

The amount of catalyst may be varied in accordance with well-known practice as, for example, from 0.01 mole to 0.5 mole of catalyst per equivalent of fatty material in the reaction.

The resulting mixture of soybean oil, glycerine, and catalyst is then reacted to cause alcoholysis of the fatty material and formation of substantial amounts of partial ester including monoglyceride. The temperature at which the reaction is carried out may vary from about 20° C. to about 300° C. depending upon the materials being treated. It is necessary, however, that the minimum reaction temperature be sufficient to cause substantial formation of the desired partial ester and it is desirable to carry out the reaction under reaction conditions effective to cause substantially maximum formation of the desired partial ester in the composition resulting from the reaction. In the case of alcoholysis of soybean oil, substantial formation of monoglyceride is initiated above about 180° C. but desirably the reaction is effected at a temperature above about 230° C. The alcoholysis of soybean oil is preferably carried out above the minimum reaction temperature at which maximum amounts of monoglyceride are obtained and desirably in the range of about 250° to 260° C., although reaction is successfully effected at a temperature of about 180°–200° C. with longer reaction times.

After reaction has been effected, the monoglyceride formed in the reaction is separated from the composition resulting therefrom while the composition contains the catalyst in active form. During the reaction, the composition reaches an equilibrium concentration between the alcohol, the convertible fatty material and the partial ester formed by the reaction. The equilibrium system reached under reaction conditions is, however, a dynamic equilibrium. A change in the relative proportions of the components in the composition causes the equilibrium to shift in order to assume a new metastable condition. Thus, for example, removable of alcohol from the composition by phase separation or other means causes reversion of partial ester to be initiated whereby additional alcohol is liberated to take the place of alcohol separated from the composition. The speed of reversion of the patrial ester is a function of the change in concentration of the alcohol, the temperature of the composition and the presence of catalyst. Elevated temperatures and presence of active catalyst in general accelerate the reversion. Lowering the temperature, however, causes alcohol to separate from the oil phase and radically changes the composition of the system.

The interrelationship of concentration, temperature, and catalyst determines the time interval necessary for substantial reversion of partial ester to be effected, and the time interval, for any given combination of conditions, which will elapse before substantial reversion of the partial ester to a more stable ester form occurs is denoted the reversion time interval.

To illustrate what is meant by reversion time interval, if soybean oil and glycerine are reacted at elevated temperatures in the presence of active catalyst, an equilibrium system is attained. If the temperature is maintained at the final reaction temperature and the concentration of the components of the system is maintained constant by preventing loss, decomposition or phase separation of the components, the system is maintained at static equilibrium for an infinite time interval.

When the system is cooled, however, glycerine separates from the reaction composition and reversion of partial ester is initiated. The lowered temperature slows the reversion and when the composition is rapidly cooled for example from a reaction temperature of 260° C. to a temperature of 100° C., the reversion time interval will be several hours. Thus, for example, 10% reversion does not occur in less than about 8 hours at 100° C.

When distillation is effected, however, to remove from the system either glycerine or partial ester or both, the reversion time interval is greatly shortened by the elevated temperatures employed. Substantial reversion of the order of 10% or more will occur in ten minutes when distillation of monoglyceride is effected from a glycerine-depleted composition at a temperature of 200° C. and at least 10% reversion will occur under distillation conditions at a temperature above about 230° C. in about 5 minutes.

I have discovered that in the process of this invention, separation of partial ester must be effected during the reversion time interval for optimum yield. Because of the shortened reversion time interval at elevated temperatures, I discovered that separation of the alcohol and partial ester must be effected simultaneously or that separation of the partial ester from a composition freed of a substantial portion of its unreacted alcohol must be effected rapidly if substantial reversion is to be obviated.

I have found that such separation of partial ester is successfully effected by spreading the composition, containing partial ester and active catalyst, in a thin film at a temperature below the decomposition temperature of the catalyst and subjecting the film to vacuum effective to distill partial ester therefrom. This is in contrast to pot type vacuum distillation where the reversion time interval is exceeded before separation is effected and objectionable reversion occurs.

Thus for optimum yield, monoglyceride present in a composition resulting from alcoholysis of soybean oil with glycerine in the presence of ester-interchange catalyst is separated from the composition, before the reversion time interval has elapsed, by vacuum distillation from a thin film leaving a distillation residue containing the catalyst in active form.

This separation is effected by maintaining the composition resulting from reaction substantially at reaction temperature and spreading successive portions on an evaporating surface maintained under vacuum so as to flash evaporate monoester and glycerine simultaneously and rapidly. Alternatively the composition is cooled to allow glycerine to separate from the composition and be removed by phase decantation whereafter the monoester and any remaining glycerine are simultaneously distilled rapidly from the glycerine-depleted composition by flash evaporation from a film as set forth hereinabove within the reversion time interval.

In the preferred separation, the separating is effected within the reversion time interval by cooling the reaction composition to about 100° C., causing a substantial proportion of the unreacted glycerine to separate from the composition, separating the layers thus obtained by phase decantation, thereafter vacuum distilling substantially all of the residual glycerine remaining in the composition therefrom by vacuum distillation of successive increments of the glycerine-depleted composition in film from under conditions ineffective to substantially distill monoglyceride therefrom, and thereafter progressively subjecting successive increments of the resulting substantially glycerine-free composition to vacuum distillation from a thin film under conditions effective to distill monoester therefrom.

Thus, separation of the desired ester is effected while the composition contains the catalyst in active form and without substantial reversion of the partial ester during the separation by spreading the composition obtained from the reaction in a thin film on an evaporating surface, maintaining the film at a temperature substantially below the decomposition temperature of the catalyst employed and subjecting the film to vacuum effective to distill monoglyceride therefrom leaving a distillation residue containing substantially all of the original catalyst still in active form. The partial ester distilled from the film is condensed and cooled substantially immediately upon being distilled from the film so that the entire separation is effected during a time interval insufficient to effect substantial reversion of the partial ester. The separation is described in greater detail in my copending application Serial No. 105,271 filed July 16, 1949.

Distillation of monogylceride or other partial ester is readily effected in this manner in a short time since, due to the thinness of the layer being distilled, substantially all the molecules of monoglyceride readily find their way to the distilling surface and escape from the heated film. Thus, no portion of the composition is subjected to objectionable local overheating nor held at an elevated temperature for an extended period of time.

Distillation is preferably effected by causing successive small increments of the composition being distilled to flow along an evaporation surface so that distillation of any given increment is completed in a time less than about five minutes. It is desirable to employ high vacuum substantially unobstructed path centrifugal distillation for effecting separation of the monoglyceride whereby relatively large quantities of monoglyceride may be distilled in a relatively short time. In the case of such high vacuum centrifugal distillation, the total distillation period for any given increment of the composition is less than a minute and usually only a few seconds. With such rapid distillation, the reversion time interval is not exceeded even with active catalyst present in the distilland. Thus the distillation of monoglyceride may be effected from a composition containing active catalyst at temperatures of the order of 130°–200° C. and distillation temperatures below about 225° C. are preferably employed.

The distillation is carried out under conditions effective to molecularly distill the monoglyceride. The pressure employed must be effective to cause distillation of the desired partial ester from the film of distilland at the temperature employed. In most cases, it is desirable to employ a vacuum below about 100 microns and preferably below about 50 microns mercury with pressures of the order of 1–20 microns or less being desirably employed.

If it is desired to obtain monoglyceride substantially free of alcohol, the unreacted glycerine in the composition resulting from the reaction is separated before vacuum distillation of the monoglyceride. Alcohols such as glycerine may be at least partially separated from the reaction mixture with fatty oils such as soybean oil by cooling and allowing the reaction composition to stand for a short time and separate into layers whereby a substantial part of the unreacted glycerine may be drawn off.

The remaining unreacted glycerine is then separated from the composition by subjecting the composition to high vacuum film distillation under conditions effective to distill off the glycerine without distilling a substantial amount of monoglyceride. The resulting glycerine-free composition is then vacuum distilled whereby monoglyceride substantially free of catalyst and containing less than 1% glycerine is obtained as distillate leaving a distillation residue comprising active catalyst and unreacted oil which consists essentially of di- and triglycerides in the case of oils such as soybean oil.

This distillation residue is then recombined with the unreacted glycerine previously separated therefrom, the resulting admixture is replenished with additional oil and glycerine and the process repeated. These successive steps are repeated until the amount of monoglyceride being recovered as distillate is substantially equivalent to the amount of convertible fatty material being added whereby substantially complete conversion of the oil to monoglyceride is effected. The active catalyst is repeatedly recycled and it is necessary only to add make-up amounts of oil and glycerine at each cycle. The amount of catalyst remains substantially constant without substantial replenishment although small amounts may be added if necessary to make up for losses which may occur during the recycling.

Thus, by means of this invention, quantity production of pure monoglyceride is made possible employing only a small initial quantity of catalyst and converting substantially all the fatty material being treated to monoglyceride and the monoglyceride recovered is substantially free of catalyst and glycerine.

Any of the partial esters prepared by the alcoholysis of fatty material may be prepared in accordance with this invention. The fatty material converted may be any of the natural or synthetic fatty acid compounds including fatty acids or fatty acid esters or mixtures thereof and including either solid or liquid fats and fatty oils. The fatty acid compounds are preferably those fatty materials having from about twelve to about twenty carbon atoms in the fatty acid chain although fatty acid chains of from about eight to about twenty-six carbon atoms are suitable for use in practising this invention. The fatty acid esters may include esters of either monohydric or polyhydric alcohols, as, for example, the fatty oils comprising a substantial proportion of triglycerides.

Suitable fatty materials which may be treated in accordance with this invention include marine oils such as fish oils and whale oil, animal fatty materials such as tallow, wool grease, hydrogenated tallow and the various other animal fats and fatty oils, and vegetable oils such as soybean oil, cottonseed oil, tung oil, coconut oil, palm oil, corn oil, olive oil, castor oil, peanut oil and the like, as well as such other fatty materials as the hydrogenated vegetable oils or shortenings. Partial esters of particularly good quality for certain uses are prepared by alcoholysis of free fatty acids such as stearic acid, oleic acid, lauric acid, palmitic acid, myristic acid and the like.

The alcohol employed in the reaction may be either monohydric or polyhydric; but where a monohydric alcohol such as methyl alcohol or ethyl alcohol is employed it is reacted with a fatty acid ester of a polyhydric alcohol to give a partial ester. Alcohols which are particularly suitable for alcoholysis are the aliphatic alcohols including the aryl-substituted alcohols and desirably the lower saturated polyhydric alcohols. Thus, suitable alcohols include the polyhydroxy alkanes such as glycerol, sorbitol, mannitol and the like; ether alcohols such as diglycerol, polyglycerols and the like; alkylene glycols such as trimethylene glycol, ethylene glycol, propylene glycol, diethylene glycol, polyethylene glycols, polypropylene glycols and the like; as well as such other alcohols as pentaerythritol and the like.

The catalyst employed in the reaction may be any ester-interchange catalyst wihch will not distill with the desired partial ester and, although the metal compounds such as alkali and alkaline earth compounds and particularly the bivalent metal compounds are preferably employed, other suitable catalysts include well-known organic and inorganic acid catalysts such as phosphoric acid, sulfuric acid or similar inorganic acid, substituted inorganic acids such as alkyl sulfuric acid, organic acids such as aryl sulphonic acids, and the like, and other acid materials, as well as such other materials as tetramethyl ammonium hydroxide, aluminum chloride, and the like. The preferred strontium compounds are the fatty acid-reactive strontium compounds and particularly the water or alcohol soluble compounds. Suitable strontium compounds include strontium oxide, strontium hydroxide, strontium chloride, strontium bromide, strontium nitrate, strontium acetate, strontium tetraborate, strontium lactate and the like and such compounds are desirably employed where product color is a prime factor.

The alcoholysis reaction is carried out at a temperature substantially above the minimum temperature necessary for formation of the desired partial ester and desirably at a temperature sufficiently above the minimum reaction temperature to establish an equilibrium system with optimum yield of the desired partial ester in a period of from ½ to 4 hours. The reaction is generally effected at temperatures of from about 20° C. to about 400° C. depending upon the materials being reacted and the reaction time desired. For example, the alcoholysis of soybean oil has been carried out at 180°–200° C. but is more preferably effected at 250°–260° or desirably at 285° C. The reaction mixture is preferably maintained at reaction temperature for about one-half hour although in some cases alcoholysis may be effected in fifteen minutes or less or the reaction may be carried on for as much as one or two hours or longer. When the reaction mixture is heated for extended periods, precautions should be taken to prevent substantial loss of the alcohol, which would shift the reaction equilibrium.

In the process according to this invention, it is desirable to preheat the reaction mixture before it is introduced into the reaction in order that the total reaction time may be minimized. The reaction mixture is preferably agitated during reaction and water evolved during the reaction should be continuously removed from the reaction zone.

The invention is particularly applicable for the preparation and recovery of monoesters such as monoglycerides but the invention may be employed in preparing any fatty acid partial ester such as diglycerides and the like. The desired partial ester may be separated and recovered with unreacted alcohol from the reaction in which case the amount of alcohol employed during the reaction may be adjusted to give a product of the desired composition.

Preferably, however, the desired partial ester is separated in substantially pure form and the unreacted alcohol is recycled with the distillation residue containing the active catalyst whereby only make-up amounts of fatty material and alcohol are added to replenish the composition from which the partial ester has been separated.

In an example of the process in accordance with this invention as represented schematically in the drawings, a fatty oil such as soybean oil is admixed with glycerine in the proportions of 2.3 moles of glycerine per mole of fatty oil calculated as triglyceride and 0.1% by weight of strontium oxide based on the weight of the reaction mixture. The strontium oxide combines with fatty acid in the oil to form a strontium soap which functions as ester-interchange catalyst in the reaction. The resulting mixture is passed through a preheater wherein the temperature is raised to about 225°–230° and the heated mixture is then led into a pot reactor. In the reactor, the mitxure is heated to 250°–260° C. and maintained at that temperature for 20 minutes to cause formation of monoglyceride. During the reaction, the mixture is constantly agitated and water vapor evolved during the reaction is vented from the reactor.

The composition resulting from the reaction, consisting primarily of active catalyst, unreacted glycerine, and mono-, di- and triglycerides, is then led through a cooling coil whereby the temperature of the composition is rapidly reduced to about 100° C.

The composition is allowed to stand without agitation for about five minutes whereupon a substantial proportion of the unreacted glycerine separates from the body of the composition and is drawn off.

The remainder of the composition which is poor in glycerine is continuously fed on to the rotor of a high vacuum substantially unobstructed path centrifugal still. The nature of centrifugal distillation is such that a single pass of a given increment of composition being distilled is usually effected in a second or two or at most in less than 2 or 3 minutes. Furthermore, with the composition spread in a thin film, the temperature of the composition can be rapidly changed. It is therefore possible to distill off glycerine in a relatively short time and at a relatively low temperature without initiating reversion of the monoglyceride.

Accordingly most of the residual glycerine remaining in the composition is separated therefrom by subjecting successive small increments of the composition to high vacuum centrifugal distillation at about 100 microns pressure and a rotor temperature of about 100° C., although pressures of 10–300 microns and temperatures of 90°–150° C. are suitable.

In order to ensure substantially pure monoglyceride being obtained as product, the distillation residue from the first distillation is immediately passed to another high vacuum unobstructed path centrifugal still whereon a second distillation is effected at about 20 microns pressure and a rotor temperature of about 120° C. whereby substantially all residual unreacted glycerine and a small amount of monoglyceride is distilled from the composition.

The resulting substantially glycerine-free distillation residue which still contains the catalyst in active form is immediately subjected to a third high vacuum centrifugal distillation at about 3 microns pressure and about 170° C. rotor temperature whereby about 30% by weight of the composition being distilled is distilled out as pure monoglyceride containing substantially no catalyst and less than 1% glycerine by weight. This distillate is immediately condensed and recovered.

The distillation residue from the third distillation is thereafter preferably molecularly distilled at a pressure of about 3 microns and a temperature of about 210° C. whereby any residual monoglyceride is stripped therefrom and returned to the system to be redistilled in the above-named third distillation. In a continuous process of successive distillations, the entire series of distillations is readily effected in less than five minutes of actual distillation time for any given increment of composition.

The final distillation residue consisting mainly of diglycerides, triglycerides and active catalyst is then recombined with the unreacted glycerine cuts separated from the reaction composition prior to recovery of the pure monoglyceride and the recombined admixture is replenished with make-up amounts of fatty oil and glycerine correlated to the amount of monoglyceride recovered and the cycle is repeated successively. Since the original catalyst is retained intact throughout the process, no additional catalyst is added. It is desirable to separate from the distillation residue small amounts of polymeric material formed in the reaction before recycling.

By the time the third cycle is being effected, the amount of monoglyceride being recovered is substantially equivalent to the amount of convertible fatty oil being added to replenish the distillation residue. Thus, the process is readily continued for extended periods of time whereby substantially all of the convertible fatty material being treated is recovered as monoglyceride.

Thus, by means of this invention it is possible to obtain substantially complete yields of monoglyceride, diglycerides or other partial ester rather than the yields of 40–50% by weight based on the weight of fatty material obtained heretofore. Furthermore, the monoglyceride obtained contains less than 1% glycerine by weight as compared to commercially available monoglyceride preparations which contain 7% or more glycerine. The monoglyceride obtained is substantially free of fatty acids, diglycerides and triglycerides rather than being mixed with the almost equal amount of diglyceride normally formed during the reaction. The distillation residue is readily recycled since it contains no materials added to inactivate the catalyst. The cost of the catalyst becomes a negligible factor since it is reused repeatedly. There is substantially no by-product to be disposed of since substantially the entire amount of fatty material is converted to the desired partial ester. The process is capable of large-scale production of monoglycerides or other partial ester of high purity which was not feasible heretofore. Substantially all the monoglyceride formed in the reaction is recovered since substantial reversion is obviated.

While the invention has been described in considerable detail with reference to the preparation and recovery of monoglyceride, it will be understood that the invention is applicable to the preparation and recovery of any fatty acid partial ester of a polyhydric alcohol or mixtures thereof and that variations and modifications may be effected within the spirit and scope of the invention in accordance with the foregoing description and drawings and as defined in the appended claims.

What I claim is:

1. The process which comprises admixing a fatty material, an alcohol and an ester-interchange catalyst, reacting the resulting mixture at a reaction temperature effective to cause substantial formation of a desired fatty acid partial ester in the composition resulting from the reaction, separating said partial ester from said composition while said composition contains said catalyst in active form, said separating being effected by spreading said composition in a thin film while maintaining said film at a temperature substantially below the decomposition temperature of said active catalyst and subjecting said composition while in said thin film to vacuum effective to distill said partial ester therefrom leaving a distillation residue containing said active catalyst, replenishing said distillation residue with make-up amounts of said fatty material and said alcohol and again effecting reaction of said replenished residue and separating partial ester formed thereby, and repeating said process at least until the amount of said partial ester being recovered is substantially equivalent to the amount of convertible fatty material being added to the distillation residue whereby substantially complete conversion of said fatty material to said partial ester is effected.

2. The process which comprises admixing a fatty material, an alcohol and an ester-interchange catalyst, reacting the resulting mixture at a temperature effective to cause substantial formation of a desired fatty acid partial ester in the composition resulting from the reaction, separating from said composition a substantial proportion of said alcohol remaining in said composition after said reaction, separating said partial ester from the remainder of said composition while said composition contains said catalyst in active form, said last-named separating being effected by spreading said composition in a thin film while maintaining said film at a temperature substantially below the decomposition temperature of said active catalyst and subjecting said film to vacuum effective to distill said partial ester from said composition leaving a distillation residue containing said catalyst in active form, recombining said distillation residue and said alcohol separated from said composition, adding thereto make-up amounts of said fatty material and said alcohol, repeating the reaction and separation of said partial ester formed thereby, and continuing the process until the amount of said partial ester being distilled from said composition is substantially equivalent to the amount of convertible fatty material being added to said distillation residue whereby substantially complete conversion of said fatty material to said partial ester is effected.

3. The process which comprises admixing fatty material with an excess of an alcohol and an ester-interchange catalyst, reacting the resulting mixture to cause maximum formation of a desired fatty acid partial ester of a polyhydric alcohol in the composition resulting from the reaction, and during the reversion time interval before substantial reversion of said partial ester is effected and while said composition contains said catalyst in active form, separating said partial ester from said composition by spreading said composition in a thin film while maintaining said composition at a temperature substantially below the decomposition temperature of said active catalyst and subjecting said film to vacuum effective to distill said partial ester therefrom leaving a distillation residue of higher ester in admixture with said active catalyst, adding to said residue make-up amounts of said fatty material and said alcohol and again reacting the resulting replenished mixture and separating said partial ester as in said first-named reaction and separation and thereafter again replenishing the distillation residue, and repeating said process until the amount of said partial ester being recovered is substantially equivalent to the amount of convertible fatty material being added where-

by substantially all of said fatty material is converted and recovered as said partial ester.

4. The process which comprises admixing fatty material with an excess of a polyhydric alcohol and a bivalent metallic ester-interchange catalyst, reacting the resulting mixture under conditions effective to establish an equilibrium composition containing substantially a maximum amount of a fatty acid monoester, recovering substantially all of said monoester present in said composition during the reversion time interval, which time interval is insufficient to permit substantial reversion of said monoester, said recovering being effected while said composition contains said catalyst in active form by first separating unreacted polyhydric alcohol from said composition and thereafter separating said monoester from said composition, said last-named separating being effected by subjecting successive portions of said composition to high-vacuum substantially unobstructed path centrifugal distillation at a temperature substantially below the decomposition temperature of said catalyst under conditions effective to distill said monoester from said composition leaving a distillation residue containing said catalyst in active form, recombining said separated unreacted polyhydric alcohol and said distillation residue, replenishing the resulting admixture with make-up amounts of said fatty material and said polyhydric alcohol, and repeating the reaction and separation of said monoester and replenishing of the distillation residue until the amount of said monoester being recovered is substantially equivalent to the amount of fatty material being added to replenish the distillation residue.

5. The process which comprises admixing a fatty oil with an excess of glycerine and an ester-interchange catalyst comprising a bivalent metal compound, reacting the resulting mixture under conditions effective to cause substantially maximum formation of monoglyceride in the composition resulting from said reaction, thereafter within a reversion time interval insufficient to permit substantial reversion of said monoglyceride and while said composition contains said catalyst in active form, separating said monoglyceride from said composition by subjecting successive portions of said composition to molecular distillation leaving a distillation residue containing said catalyst in active form, replenishing said distillation residue with additional fatty oil and glycerine, and repeating the reacting, separating and replenishing a plurality of times whereby the amount of additional fatty oil being added is substantially equivalent to the amount of monoglyceride being separated from the composition resulting from the reaction.

6. The process which comprises converting substantially all of the fatty constituents of a vegetable oil to monoglyceride, said converting being effected by reacting a predetermined amount of said vegetable oil with an excess of glycerine in the presence of a bivalent metal soap catalyst at a temperature effective to cause substantial formation of monoglyceride in the composition resulting from the reaction, separating said monoglyceride from said composition while said composition contains said catalyst in active form, said separating being effected by subjecting successive portions of said composition to high vacuum unobstructed path centrifugal distillation at a temperature substantially below the decomposition temperature of said catalyst under conditions effective to distill monoglyceride from said composition leaving a distillation residue containing said catalyst in active form, replenishing said distillation residue with additional vegetable oil and glycerine, and repeating said process successively until the amount of monoglyceride being separated is substantially equivalent to the amount of vegetable oil being added to the distillation residue.

7. The process which comprises successively and repeatedly reacting fatty material and polyhydric alcohol in the presence of an ester-interchange catalyst under conditions causing substantial formation of a monoester in the composition resulting from the reaction, separating said monoester from said composition, while the catalyst is in active form, by molecularly distilling said monoester from said composition leaving a distillation residue containing said active catalyst, and replenishing said distillation residue with additional fatty material and polyhydric alcohol, said process being continued at least until the amount of said monoester being distilled from said composition is substantially equivalent to the amount of said fatty material being added to said residue.

8. The process which comprises the steps of reacting a fatty acid compound with a polyhydric alcohol in the presence of a bivalent metal soap catalyst under conditions effective to cause substantial formation of monoester in the composition resulting from the reaction, separating said monoester and unreacted polyhydric alcohol from said composition and from each other, while said composition contains said catalyst in active form by repeated high vacuum substantially unobstructed path centrifugal distillation under conditions leaving a distillation residue substantially free of polyhydric alcohol and monoester but containing said catalyst in active form, recombining said separated unreacted polyhydric alcohol with said distillation residue, replenishing the resulting mixture with additional fatty acid compound and polyhydric alcohol, and successively repeating said steps at least until the amount of said monoester being separated from said composition is substantially equivalent to the amount of fatty acid compound being added to replenish said distillation residue.

9. The process which comprises repeating the successive steps of reacting a fatty acid compound with glycerine in the presence of a bivalent metal soap catalyst under conditions effective to cause substantially maximum formation of monoglyceride in the composition resulting from the reaction, recovering substantially pure monoglyceride formed in said reaction by separating said monoglyceride from said composition during a time not substantially greater than the reversion time interval, which reversion time interval is insufficient to permit substantial reversion of said monoglyceride, and while said composition contains said catalyst in active form leaving a residue containing said active catalyst, said separating being effected by first treating said composition to remove therefrom substantially all unreacted glycerine, said treating including subjecting successive portions of said composition to high vacuum unobstructed centrifugal distillation under conditions causing substantially all of said glycerine to distill from said composition without distilling a substantial amount of monoglyceride therefrom, and thereafter subjecting successive portions of said composition substantially freed of glycerine to high vacuum substantially unobstructed path centrifugal distillation

under conditions effective to distill substantially pure monoglyceride therefrom at a temperature substantially below the decomposition temperature of said active catalyst, recombining said residue containing said active catalyst with said unreacted glycerine removed therefrom, and replenishing the resulting recombined mixture with additional fatty acid compound and glycerine.

10. The process of converting fatty material to fatty acid partial ester of polyhydric alcohol which comprises establishing a cyclic system by repeatedly in successive stages of said system effecting ester-interchange in a reaction mixture between fatty material and an alcohol in the presence of an ester-interchange catalyst under conditions causing substantial formation of a fatty acid partial ester of polyhydric alcohol in the composition resulting from the reaction, separating said partial ester from the remainder of said composition while said catalyst is in active form by progressively spreading said composition in a thin film and vacuum distilling said partial ester from said composition in said thin film leaving a distillation residue containing said catalyst in active form, and recycling said distillation residue to again effect ester-interchange in the reaction mixture, progressively withdrawing from said cyclic system partial ester formed in said reaction mixture and progressively adding to said cyclic system fatty material and alcohol in amounts substantially equivalent to said partial ester being withdrawn from said system.

11. In the process of converting fatty material to a fatty acid partial ester of a polyhydric alcohol by reacting fatty material with an alcohol in the presence of an ester-interchange catalyst, the method which comprises establishing a continuously circulating system containing said ester-interchange catalyst in active condition throughout said system, progressively adding to said system reacting proportions of said fatty material and said alcohol, effecting an ester-interchange reaction between said fatty material and said alcohol in the course of said circulating in said system, and progressively removing from said system partial ester produced in said reaction, said removing being effected by progressively spreading the composition resulting from said reaction in a thin film and vacuum distilling said partial ester from said composition leaving a distillation residue containing said catalyst in active form, coordinating the quantities of fatty material and alcohol progressively added to said system with the amount of partial ester progressively removed from said system and thereby maintaining predetermined equilibrium conditions in said system, the quantity of catalyst in said system remaining substantially constant without substantial replenishment.

12. The process which comprises repeatedly and successively subjecting a reaction mixture comprising fatty material in admixture with a polyhydric alcohol and a strontium soap ester-interchange catalyst to an alcoholysis reaction, separating from said reaction mixture polyhydric alcohol fatty acid partial ester formed in said reaction, and adding to said reaction mixture reactive amounts of fatty material and polyhydric alcohol, said strontium soap catalyst being in active form throughout said process, said separating being effected following said reaction by progressively spreading said reaction mixture in a thin film on a vaporizing surface, maintaining said film at a temperature substantially below the decomposition temperature of said strontium soap catalyst, and subjecting said reaction mixture while in said thin film to vacuum effective at said temperature to distill said partial ester from said reaction mixture.

13. The method of preparing a monoester composition substantially free of objectionable color contaminants, which method comprises establishing a cyclic system having an ester-interchange catalyst comprising a strontium compound maintained in active form throughout said system, said cyclic system comprising repeating the successive steps of (1) effecting reaction in a reaction zone between a fatty material and an alcohol in the presence of said catalyst under conditions effective to convert a substantial proportion of said fatty material to a fatty acid monoester of a polyhydric alcohol, (2) separating monoester formed in said reaction from the mixture resulting from said reaction, said separating being effected while said catalyst is maintained in active form by progressively spreading the mixture resulting from said reaction in a thin film and vacuum distilling said monoester therefrom, and (3) returning said mixture from which said monoester has been separated to said reaction zone, withdrawing separated monoester from said system and adding to said system fatty material and alcohol in amounts correlated to the amount of monoester withdrawn.

14. The cyclic process for preparing monoglyceride of uniform color which comprises establishing a cyclic system comprising the successive steps of (1) reacting in a reaction zone fatty material including a substantial proportion of triglyceride with glycerine in the presence of a strontium soap ester-interchange catalyst, (2) separating in a separating zone monoglyceride formed in the reaction from the composition resulting from said reacting while said composition contains said catalyst in active form, said separating being effected by progressively spreading said composition in a thin film and vacuum distilling monoglyceride from said composition, and (3) returning the resulting monoglyceride-depleted composition containing said active catalyst to said reaction zone, withdrawing from said system monoglyceride separated in said separating zone and adding to said system fatty material and glycerine in amounts correlated to the monoglyceride being withdrawn from said system and effective to maintain predetermined equilibrium conditions in said system.

15. The method of producing monoglycerides which comprises reacting triglyceride fatty material with glycerine in the presence of a metal soap catalyst and thereby forming a composition containing a substantial amount of monoglyceride, separating monoglyceride from the remainder of said composition while said composition contains said catalyst in active form by subjecting said composition to thin film high vacuum distillation, recycling said remainder of said composition including said catalyst in active form, adding to the recycling remainder of said composition amounts of triglyceride fatty material and glycerine substantially equivalent to said monoglyceride separated from said composition, and repeating said reacting, separating, recycling and adding.

16. The process of preparing a monoglyceride product which comprises establishing a continuously circulating system containing ester-interchange catalyst maintained in active form

throughout said system, effecting alcoholysis of fatty material with glycerine in the course of said circulating system, and adding fatty material and glycerine to said circulating system and vacuum distilling monoglyceride from said system in correlated amounts effective to maintain predetermined equilibrium conditions in said system, said vacuum distilling being effected from a thin film at a pressure below 100 microns Hg.

17. The process which comprises the sequential steps of reacting a mixture of fatty acid and glycerine in the presence of an ester-interchange catalyst and thereby forming a composition containing a substantial amount of monoglyceride, separating unreacted glycerine from said composition, progressively spreading the resulting glycerine-depleted composition in a thin film and vacuum distilling monoglyceride from said composition while said catalyst is in said composition in active form and leaving a distillation residue containing said active catalyst, and replenishing said distillation residue with additional fatty acid and glycerine in amounts correlated to the amount of monoglyceride distilled from said composition and thereby forming a reconstituted mixture for said reacting, and repeating said reacting and said vacuum distilling with said reconstituted mixture.

18. The process which comprises the sequential steps of reacting a mixture of fatty material and glycerine in the presence of an ester-interchange catalyst and thereby forming a composition containing a substantial amount of monoglyceride, separating a substantial portion of the unreacted glycerine from said composition by phase separation, vacuum distilling substantially all of the remaining unreacted glycerine from said composition without distilling off substantial amounts of monoglyceride, and thereafter progressively spreading said composition in a thin film and vacuum distilling monoglyceride from said composition while said catalyst is in said composition in active form and leaving a distillation residue containing said active catalyst, and replenishing said distillation residue with additional fatty material and glycerine in amounts correlated to the amount of monoglyceride distilled from said composition and thereby forming a reconstituted mixture for said reacting, and repeating said sequential steps with the reconstituted mixture at least until the amount of fatty material and glycerine added to replenish the distillation residue is substantially equivalent to the amount of monoglyceride distilled from the composition resulting from the reacting.

19. The process which comprises the sequential steps of reacting a mixture of vegetable oil and glycerine in the presence of a strontium soap catalyst and thereby forming a composition containing a substantial amount of monoglyceride, separating unreacted glycerine from said composition, progressively spreading said composition in a thin film and vacuum distilling monoglyceride from said composition while said composition contains said catalyst in active form and leaving a distillation residue containing said active catalyst, and replenishing said distillation residue with additional vegetable oil and glycerine in an amount substantially equivalent to the amount of monoglyceride distilled from said composition and thereby forming a reconstituted mixture for said reacting, and repeating said sequential steps with said reconstituted mixture.

NOEL H. KUHRT.

REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
2,496,328	Bell et al. ----------	Feb. 7, 1950

LIBRARY
FEB 26 1947
U. S. PATENT OFFICE

Concise Chemical and Technical *DICTIONARY*

Edited by

H. BENNETT

Technical Director, Glyco Products Co., Inc.
Fellow American Institute of Chemists; Fellow American Association for the Advancement of Science; Member of American Chemical Society, Society of Chemical Industry, American Association of Textile Chemists and Colorists, American Institute of Testing Materials; Editor-in-Chief: *The Chemical Formulary; Practical Emulsions; Commercial Waxes; etc.*

DEFENDANT'S
EXHIBIT
2

1 9 4 7
CHEMICAL PUBLISHING COMPANY, Inc.
BROOKLYN — NEW YORK

glycerite. Solution of medicaments in glycerin.

glycerite of boroglycerin. See boroglycerin glycerite.

glycero carbolic acid (glycerino carbolic acid). $C_3H_5(OH)_2C_6H_5O$; m.w. 168.18; col. liq.; s.w.; s.al.; used as disinfectant.

Glycerogen. Glycerin substitute consisting of glycerin, hexitols, dihydric alcohols and water.

glycerogenic. See steatolytic.

glycerol (glycerin; 1,2,3-propanetriol; trihydroxy propane; glycyl alcohol; propenyl alcohol). $CH_2OHCHOHCH_2OH$; m.w. 92.06; col. liq. or rhomb.; m.p. 17.9; b.p. 220; s.w.; s.al.; used as solvent, in medicine, perfumery, antifreeze, in mfg. soaps, explosives, etc.

glycerol borate (glyceryl borate). $(C_3H_5BO_3)_x$; m.w. $(99.86)_x$; yel. glassy.

glycerol α-chlorohydrin. See 3-chloro-1,2-propanediol.

glycerol diacetate (diacetin). $C_3H_5(OH)(OOCCH_3)_2$; m.w. 176.09; col. liq.; m.p. 40; b.p. 176^{40}; s.w.; s.al.

glycerol 1,3-dilaurate (α,γ-dilaurin). $(C_{11}H_{23}COO)_2C_3H_5OH$; m.w. 456.41; cr.; m.p. 56.6; s.al.

glycerol 1,3-dinitrate. $C_3H_5(OH)(NO_3)_2 \cdot \frac{1}{2}H_2O$; m.w. 191.07; liq.; m.p. <−30; b.p. 148^{15}; s.al.

glycerol 1,3-dipalmitate (α,γ-dipalmitin). $(C_{15}H_{31}COO)_2C_3H_5OH$; m.w. 568.53; cr.f.al. or chl.; m.p. 70; s.al.

glycerol 1,3-distearate (α,γ-distearin). $(C_{17}H_{35}COO)_2C_3H_5OH$; m.w. 624.59; rhomb. pl. f. chl. or lgr.; m.p. 79.1; s.al.

glycerol ether (of Berthelot and de Luca) (glyceryl ether). $C_6H_{10}O_3$; m.w. 130.08; col. liq.; b.p. 173; s.w.; s.al.

glycerol ethylidene ether. See acetoglyceral.

glycerol monoacetate (monoacetin; acetin). $CH_2OHCHOHCH_2OOCCH_3$; m.w. 134.08; col. oil; sp.gr. $1.206^{20/4}$; b.p. 158^{165}; s.w.; s.al.; used in mfg. dynamite, tanning.

glycerol monobutyrate (monobutyrin; glyceryl monobutyrate). $C_3H_7COOCH_2CH(OH)CH_2OH$; m.w. 162.11; col. liq.; sp.gr. 1.088; b.p. 269; s.w.

glycerol *l*-monolaurate (α-monolaurin). $C_{11}H_{23}COOCH_2CHOHCH_2OH$; m.w. 274.23; wh. need.; m.p. 63; s.al.

glycerol α-mononitrate. $CH_2OHCHOHCH_2ONO_2$; m.w. 137.06; col. pr.; m.p. 58; b.p. 155–60; s.w.; s.al.

glycerol β-mononitrate. $CH_2OHCH(ONO_2)CH_2OH$; m.w. 137.06; leaf.; m.p. 54; b.p. 160; s.w.; s.al.

glycerol *l*-monoöleate (monoölein). $C_{17}H_{33}COOCH_2CHOHCH_2OH$; m.w. 354.30; m.p. 35; i.w.; s.al.

glycerol *l*-monopalmitate (α-monopalmitin). $C_{15}H_{31}COOCH_2CHOHCH_2OH$; m.w. 330.30; leaf.; m.p. 77.

glycerol monoricinoleate (glyceryl monoricinoleate; ricinoleyl glycerin). $C_{17}H_{32}COOC_3H_5(OH)_2$; m.w. 356.31; amber liq.; s.al.

glycerol *l*-monostearate (α-monostearin). $C_{17}H_{35}COOCH_2CHOHCH_2OH$; m.w. 358.33; need. or waxlike sld.; m.p. 81.1; s.al.

glycerol *l*-octadecyl ether. See 3-octadecyloxy 1,2-propanediol.

glycerol phosphoric acid. See glycerophosphoric acid.

glycerol triacetate (triacetin). $C_3H_5(OOCCH_3)_3$; m.w. 218.11; col. liq.; m.p. −78; b.p. 259; s.al.

glycerol tribenzoate (tribenzoin). $C_3H_5(OOCC_6H_5)_3$; m.w. 404.16; need. f.me.al.; m.p. 76.5; i.w.; s.al.

glycerol tribromohydrin. See 1,2,3-tribromo propane.

glycerol tributyrate (butyrin; tributyrin). $(CH_3CH_2CH_2CO)_3C_3H_5O_3$; m.w. 302.20; col. oily liq.; m.p. <−75; b.p. 315; i.w.; s.al.

glycerol trichlorohydrin. See 1,2,3-trichloro propane.

glycerol trilaurate (trilaurin; laurin). $(C_{11}H_{23}COO)_3C_3H_5$; m.w. 638.58; col. need.; m.p. 46.4; i.w.; s.al.

glycerol trimyristate (myristin; trimyristin). $(C_{13}H_{27}COO)_3C_3H_5$; m.w. 722.67; glit. need. f.et.; m.p. 56.5; i.w.; s.al.

glycerol trinitrate. See nitroglycerin.

glycerol trinitrite. $C_3H_5(ONO)_3$; m.w. 179.06; yel. liq.; i.w.

glycerol trioleate (triolein; olein; glyceryl oleate). $(C_{17}H_{33}COO)_3C_3H_5$; m.w. 884.81; col. oil; m.p. −17; b.p. 240^{18}; i.w.; s.al.

glycerol tripalmitate (tripalmitin; palmitin). $(C_{15}H_{31}COO)_3C_3H_5$; m.w. 806.76; col. need. f.et.; m.p. 65.1; b.p. 310–20; i.w.

glycerol tristearate (stearin; tristearin). $(C_{17}H_{35}COO)_3C_3H_5$; m.w. 890.86; col. cr. f.et.; m.p. 54.5; i.w.; s.al.

glycero phosphate. Salt of glycerophosphoric acid; e.g., calcium glycerophosphate, $CaC_3H_5(OH)_2PO_4$; used in medicine.

glycero phosphoric acid (glycerol-phosphoric acid; glycerino phosphoric acid). $C_3H_5(OH)_2OPO_3H_2$; m.w. 172.09; col. oily liq.; m.p. −20; s.w.; s.al.

glyceryl benzoate (tribenzoin). $CH_2OOCC_6H_5CHOOCC_6H_5CH_2OOCC_6H_5$; m.w.

PHYSICAL CHEMISTRY FOR COLLEGES

A COURSE OF INSTRUCTION BASED UPON
THE FUNDAMENTAL LAWS OF CHEMISTRY

BY

E. B. MILLARD
ASSISTANT PROFESSOR OF PHYSICAL CHEMISTRY
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

DEFENDANT'S
EXHIBIT
3

FIRST EDITION

McGRAW-HILL BOOK COMPANY, INC.
NEW YORK: 370 SEVENTH AVENUE
LONDON: 6 & 8 BOUVERIE ST., E. C. 4
1921

CHAPTER IV

SOLID SUBSTANCES

The most important characteristics of the solid state are rigidity, and a definite arrangement of the atoms or molecules of a crystal with reference to one another. It has long been known that the forces acting between constituent atoms of solid substances were specific in character, and of great magnitude. In general there is no tendency for a substance in crystallizing from its melt or from solution to include the molecules of other substances; and this affords a ready means of purification of the products of chemical operations. Certain chemically related substances have the property of forming mixed crystals; in these cases recrystallization is not a suitable method of purification, and other means suited to the problem in hand must be employed.

The changes in properties attending the passage of a substance from liquid to solid are much smaller than those which attend the condensation of a gas. The density of a solid is commonly within 10 per cent of that of its liquid, while the density of a liquid is often a thousand times that of the vapor from which it was formed. Solids have vapor pressures, just as liquids have; they increase with increasing temperature as in the case of liquids. The rate at which the vapor pressure, or sublimation pressure, changes with temperature may be calculated from the Clausius-Clapeyron equation; but since the change in state is from solid to vapor, the heat term is the sublimation heat of the solid. The equation is

$$\frac{dp}{dT} = \frac{L_s}{T\Delta v} = \frac{pL_s}{RT^2}$$

where L_s is the molecular latent heat of sublimation, and the vapor formed has the volume RT/p.

The melting point of a substance is that temperature at which its liquid and solid phases are in equilibrium under a pressure of 1 atmosphere. As the presence of some other substance lowers the melting point in proportion to the quantity of it present, an accurate determination of melting point is a ready means of establishing the purity of a product.

Changes of pressure have but slight effect upon the melting point of a solid, except where very high pressures are involved. For example, an increase of 2,100 atmospheres lowers the melting point of ice 22°. The effect of pressure can be calculated from the Clausius-Clapeyron equation,

$$\frac{dp}{dT} = \frac{L_f}{T\Delta v}$$

where Δv is the increase of volume which takes place on melting, and L_f is the heat of fusion. Here the approximate form of this equation obtained from the ideal gas law cannot of course be applied. If the effect of pressure is desired in degrees per atmosphere, Δv should be expressed in cubic centimeters, and L_f in cubic-centimeter-atmospheres. One calorie is equivalent to 41.24 c.c. atmospheres. For example, the heat absorbed during the melting of a gram of water is 79 cal., the increase in volume upon melting is -0.09 c.c., whence

$$\frac{dp}{dT} = \frac{79 \times 41.24}{-0.09 \times 273} = -132$$

atmospheres per degree, which corresponds to $\dfrac{1}{-132} = -0.0076$ degrees per atmosphere. The negative sign indicates that pressure lowers the melting point of ice.

By considering the kinetic energy of the molecules of solids, and their energy of vibration, Honda[1] has derived an approximate relation between the heat of fusion and absolute melting point, which is somewhat similar to Trouton's rule for liquids. The rule is restricted to elements alone, and states that the atomic heat of fusion in small calories is two times the absolute melting point. The rule holds roughly for several metals, but is greatly in error for many others, indicating that the true relation between latent heat and melting point has not yet been discovered. Thus the quotient of L/T is 4.8 for bromine and chlorine, 3.3 for tin, 4.7 for bismuth, 3.8 for iodine, etc., but between 1.9 and 2.6 for most metals of high melting point.

---

[1] *Physical Review*, **12**, 425 (1918).

MELTING and SOLIDIFICATION of FATS

DEFENDANT'S EXHIBIT
4 FOR

ALTON E. BAILEY
VOTATOR DIVISION
THE GIRDLER CORPORATION
LOUISVILLE, KENTUCKY

LIBRARY
MAR 10 1952
U S PATENT OFFICE

1950
INTERSCIENCE PUBLISHERS, INC., NEW YORK
INTERSCIENCE PUBLISHERS LTD., LONDON

The approximate Clausius-Clapeyron equation for the liquid has the form:

$$d \ln p_L/dT = (H_V - H_L)/RT^2 \qquad (12)$$

where p_L refers to the vapor pressure of the liquid and $(H_V - H_s)$ is the difference in heat contents of vapor and liquid. For the solid the corresponding equation is:

$$d \ln p_S/dT = (H_V - H_S)/RT^2 \qquad (13)$$

where p_s is the solid vapor pressure and $(H_V - H_s)$ is the difference in heat contents of vapor and solid.

3. The Ideal Solution

From the preceding equations there can be developed certain useful relationships applicable to melting and solidification in either mixtures or solutions. First, however, it is neccessary to define the important concept of the perfect or *ideal solution.* An ideal solution exists when a liquid contains different molecular species which are indifferent to each other in the sense that attraction between molecules of A is no greater and no less than attraction between molecules of B, or the attraction between molecules of A and B. When this condition is fulfilled the presence of either substance can have no effect on the other except that of a simple dilution. The presence of B, for example, serves to diminish the vapor pressure of A merely to the extent that it reduces the molar concentration of B. In an ideal solution the vapor pressure of any component A is defined by *Raoult's law*:

$$p_A = p_A^{\circ} N_A$$

where p_A is the partial vapor pressure of A in the solution, p_A° is the vapor pressure of pure A at the same temperature and N_A is the mole fraction of A in the solution.

Ideal solutions are, of course, seldom or never encountered in practice. Very often, however, two substances are sufficiently similar that any slight deviation from ideality is not important, and, in any case, deviations are best handled by proceeding from the simplified case of ideality.

4. Calculation of Ideal Solubility

It is well known that the melting point of a substance is usually lowered by the admixture of a small proportion of any second substance, even if the latter is relatively high melting. The reason for this will be apparent from reference again to Figure 18. For the melting point of the substance in this diagram to be reduced from T_m to T', it is only necessary for it

INDUSTRIAL
OIL AND FAT PRODUCTS

DEFENDANT'S
EXHIBIT
5

ALTON E. BAILEY
SENIOR CHEMICAL TECHNOLOGIST,
SOUTHERN REGIONAL RESEARCH
LABORATORY, U. S. DEPARTMENT OF
AGRICULTURE, NEW ORLEANS, LOUISIANA

LIBRARY

DEC 6 1946

U. S. PATENT OFFICE

1 9 4 5
INTERSCIENCE PUBLISHERS, INC. — NEW YORK, N. Y.

the oils. Copper, in particular, is a very strong pro-oxidant,[15] being effective in a concentration of less than one part per million. Copper and copper-bearing alloys are consequently avoided in the fabrication of equipment fo handling edible fats and oils; the use even of brass valves or cocks is avoide in mills and refineries. From a practical standpoint, the possible har which a metal may do to the stability of oils depends both upon its pro-oxidant activity *per se*, and the extent to which it may become dissolve in the oil. Materials high in free fatty acids are much more inclined t dissolve metals than are neutral oils. In ordinary processing operatio common carbon steel does not appear to be sufficiently corroded by oils t seriously affect their stability.

The stability of fats is usually evaluated in the laboratory by subjecti samples to conditions which greatly accelerate their normal course of oxidation, and noting the time required for rancidity to develop. In the *aeratio test*, otherwise known as the *Swift stability test*,[15] samples are aerated a 97.7°C. In a modification of this test,[16] the evaluation of stability is mad even more rapid by conducting the aeration at 110°C. The *Schaal tes* involves simple incubation of the sample in an oven at 60°C.

(*f*) *Reactions of Hydroxyl Groups*

Hydroxyl groups may be introduced into fatty acid chains by variou means; in the case of the castor oil, which contains ricinoleic acid, they a present naturally. In industrial practice, the most important reactio involving the hydroxyl group is the dehydration of castor oil to produce drying oil with conjugated acids. This process is discussed on pag 696–697.

Hydroxyl groups in fats are readily acetylated by reaction with acety chloride or acetic anhydride in the presence of pyridine, etc.; this reactio constitutes the basis of various analytical methods for the estimation o these groups. The *acetyl value* is defined as the number of milligrams o potassium hydroxide required to neutralize the acetic acid produced b hydrolysis of one gram of acetylated fat. The *hydroxyl number* is th number of milligrams of potassium hydroxide equivalent to the hydroxy content of one gram of fat. Methods for the estimation of hydroxy content do not differentiate between hydroxyl groups in the fatty ac chains and those present in mono- or diglycerides, hence partially hydrolyzed fats may exhibit a considerable acetyl value without containi hydroxy acids.

---

[15] A. E. King, H. L. Roschen, and W. H. Irwin, *Oil & Soap*, **10**, 204–207 (1933).
[16] V. C. Mehlenbacher, *Oil & Soap*, **19**, 137–139 (1942).
[17] N. T. Joyner and J. E. McIntyre, *Oil & Soap*, **15**, 184–186 (1938).

BRIEF FOR APPELLANT

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 19,275

WOLFGANG BAENITZ, and
CHEMISCHE WERKE WITTEN,
Appellants,

v.

DAVID M. LADD,
Commissioner of Patents,
Appellee

Appeal From the United States District Court
For the District of Columbia

United States Court of Appeals
for the District of Columbia Circuit

FILED JUN 11 1965

Nathan J. Paulson
CLERK

PAUL M. CRAIG, JR.
Pennsylvania Building
Washington, D. C.

Attorney for Appellants

DICKE & CRAIG
Pennsylvania Building
Washington, D. C.

Of Counsel

STATEMENT OF QUESTIONS PRESENTED

The questions presented to this Court for decision are:

1. Whether in a trial by the Court, it was proper for the District Court to rely, as material evidence, on documentary evidence neither presented nor received at trial but only argued in the Brief of Defendant-Appellee submitted after trial;

2. Whether in the absence of any evidence whatsoever, it was proper on the part of the District Court to assume that Christensen used 80% crude glycerine in his reaction and Appellant Baenitz used 100% pure glycerine in his reaction;

3. Whether there was sufficient evidence for a finding by the Court that Plaintiff-Appellant's claimed process range overlaps the range disclosed in the Christensen patent;

4. Whether there was sufficient evidence for a finding by the Court that the processes and inherently resulting products of Christensen and of Plaintiff-Appellant Baenitz are essentially the same;

5. Whether there was sufficient evidence for the Court's holding that the control of the melting point as claimed in the process of Plaintiff-Appellant Baenitz, would have been obvious to a person skilled in the art having all those disclosures before him which were properly introduced into evidence and received as evidence at the trial, bearing also in mind the definition of the term "process" as defined in Title 35 of the U. S. Code, Section 100(b);

6. Whether it was proper for the Court to deny Plaintiff-Appellant's Motion to Vacate Judgment under Rule 59 FRCP since such denial was based (a) on reliance on evidence not properly before the

Court, (b) on a misinterpretation of the applicable statutes, and (c) on a misconception of the nature of the proceedings involved;

7. Whether the "clearly erroneous" test applied by the District Court as regards the burden of proof of Plaintiffs-Appellants was proper in proceedings constituting a trial de novo.

INDEX

JURISDICTIONAL STATEMENT 1
STATEMENT OF THE CASE 2
1. History of Case 2
2. The Invention 3
3. The Chemistry Involved 4
4. Evidence at Trial 6
STATUTES INVOLVED 10
STATEMENT OF POINTS 12
SUMMARY OF ARGUMENT 13
ARGUMENT 16
1. Reliance on Improper Evidence 16
2. Failure to Grant Motion To Vacate Judgment 19
3. Insufficiency of Additional Evidence 20
4. Identity of Christensen and Baenitz Process and Products 24
5. Obviousness of Invention 28
6. Burden of Proof 33
CONCLUSION 34

TABLE OF CASES

Douglas Aircraft Co. v. Mueller, 107 U.S.App.D.C. 321, 277 F.2d 351 18

Esso Standard Oil Co. v. Sun Oil Co., 97 App. D.C. 154, 229 F.2d 37 33

Johns-Manville Corporation v. David L. Ladd, 117 App. D.C. 262, 328 F.2d 563 34

Kay et al. v. Cain, 81 U.S. App. D.C. 24, 154 F.2d 205 18

Zenith Corporation v. Ladd, 114 App. D.C. 54, 310 F.2d 859 33

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 19,275

WOLFGANG BAENITZ, and
CHEMISCHE WERKE WITTEN,
Appellants,

v.

DAVID M. LADD,
Commissioner of Patents,
Appellee

*Appeal From the United States District Court
For the District of Columbia*

JURISDICTIONAL STATEMENT

Jurisdiction of the lower court is based on U. S. Code, Title 35, Section 145.

Jurisdiction of this Court is based on U. S. Code, Title 28, Section 1291.

STATEMENT OF CASE

1. History of Case

Appellant, Wolfgang Baenitz, filed an application for patent in the United States Patent Office on February 12, 1959, entitled "Production of Fats," which was given Serial No. 792,706 (JA 2, 6, and 43 through 47). The claims of this application, which are directed to a "method for controlling the melting point of hard and brittle fats within a relatively narrow desired range during the manufacture thereof" and evolved in the course of prosecution before the U. S. Patent Office into claims 25 to 30, were finally rejected by the Examiner of the U. S. Patent Office, and such rejection was sustained by the Board of Appeals of the U. S. Patent Office (JA 48 through 51).

A civil action to obtain Letters Patent was duly filed on June 27, 1963 (JA 1 through 5). The case came on for trial on June 1 and June 2, 1964 (JA 8 and 34).

In line with the Court's suggestion at the end of the trial on June 2, 1964, Plaintiffs-Appellants and Defendant-Appellee submitted Briefs after Trial.

In an opinion filed January 4, 1965, the District Court found for the Defendant-Appellee and against Plaintiffs-Appellants, dismissing the Complaint and in an Order of the same date ordered that judgment be entered in favor of the Defendant-Appellee (JA 29-34).

A Motion to Vacate Judgment under Rule 59 F.R.C.P. was filed by Plaintiffs-Appellants on January 14, 1965 (JA 35 through 41). In an Order filed January 28, 1965, the District Court, having found that a technical inaccuracy appears in its original opinion, modified its original opinion, but denied Plaintiffs-Appellants' Motion to Vacate Judgment and held that the said inaccuracy is insufficient, in view of the evidence as a whole, to alter the Court's ultimate determination.

Notice of Appeal to this Court was filed on February 26, 1965.

2. The Invention

The invention described in the Baenitz application relates to a process for producing hard and brittle fats by esterifying a saturated fatty acid having between 10 and 18 carbon atoms per molecule (JA 30, 43).

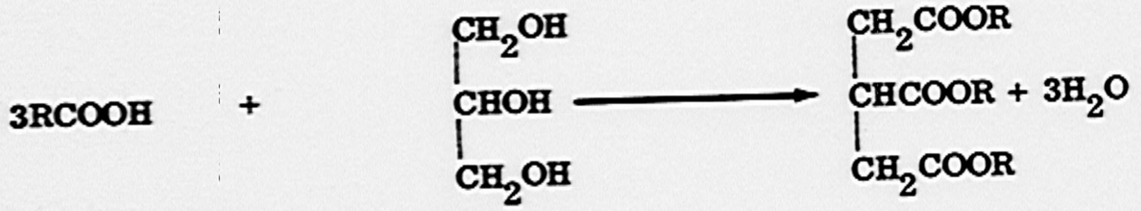
Such brittle and hard fats are especially required for use in the pharmaceutical industry as suppository bases or masses. As is well known, suppositories and thus the suppository masses must have an accurately determined melting point so as to melt at the body temperature, i.e., when in contact with the human body. On the other hand, for convenience of use, the suppository masses or bases are also required to be hard and brittle at temperatures just below the intended melting temperature, i.e., the normal temperature of the human body.

The invention described in the Baenitz application relates in particular to a method for controlling directly and accurately the melting point of hard and brittle fats as may be used for suppository bases or masses within a relatively narrow desired range during the very step of the esterification of the selected fatty acid or acids with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100 (JA 30, 44, 45). Appellant Baenitz discovered that in esterifying the saturated fatty acids having about 10 to 18 carbon atoms per molecule and mixtures thereof with such an excess of glycerine as will produce an ester mixture having a hydroxyl number between about 40 and 100, the melting point of the suppository mass or base constituted by the resulting ester mixture cannot only be accurately controlled but can even be lowered compared to the known melting points of the corresponding triglycerides, diglycerides and monoglycerides (the Table at the bottom of JA 45; JA 52b and 52d).

3. The Chemistry Involved

The general possibility of esterification of fatty acids by the reaction with alcohols such as glycerine (or glycerol) was well known for quite some time prior to Appellant's invention; however, Appellant's invention and discovery as defined by the claims of the involved repatent application is not concerned with these broad principles but relates to a process of accurately controlling the melting point of hard and brittle fats during the esterification thereof, i.e., during the reaction, generally known heretofore. The **theoretical** equations for obtaining triglycerides, diglycerides, and monoglycerides, known before the invention of Appellant Baenitz are as follows:

a) to produce a full ester or *triglyceride:*

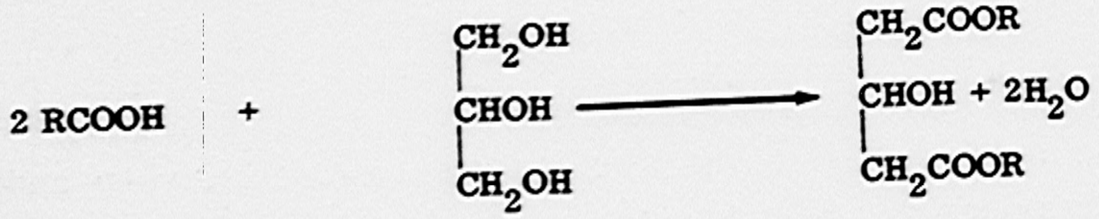


$$3RCOOH + \begin{array}{l} CH_2OH \\ | \\ CHOH \\ | \\ CH_2OH \end{array} \longrightarrow \begin{array}{l} CH_2COOR \\ | \\ CHCOOR \\ | \\ CH_2COOR \end{array} + 3H_2O$$

(3 moles of fatty acid) (1 mole of glycerine) (triglyceride)

Thus, a triglyceride is obtained when the stoichiometric amount (amount necessary for complete reaction of all the functional reactive chemical groups) of glycerine is used.

b) to produce a *diglyceride,* a partial ester:

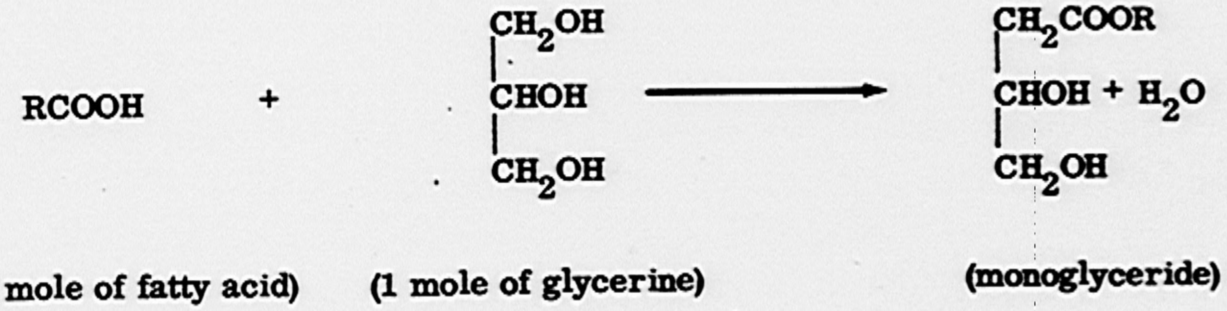


$$2\ RCOOH + \begin{array}{l} CH_2OH \\ | \\ CHOH \\ | \\ CH_2OH \end{array} \longrightarrow \begin{array}{l} CH_2COOR \\ | \\ CHOH \\ | \\ CH_2COOR \end{array} + 2H_2O$$

(2 moles of fatty acid) (1 mole of glycerine) (diglyceride)

Thus, the use of an excess amount of glycerine, i.e., an excess over the stoichiometric amount needed to form a full ester, gives a partial ester — a diglyceride in this case — because there are more hydroxyl groups available (on the glycerine) than there are carboxyl groups (on the fatty acid) with which to react.

c) to produce a *monoglyceride,* a partial ester:



$$RCOOH + \begin{array}{l} CH_2OH \\ | \\ CHOH \\ | \\ CH_2OH \end{array} \longrightarrow \begin{array}{l} CH_2COOR \\ | \\ CHOH + H_2O \\ | \\ CH_2OH \end{array}$$

Thus, the use of an even greater excess amount of glycerine over the stoichiometric amount needed to form a full ester gives a monoglyceride partial ester, again because there are three hydroxyl groups (per glycerine molecule) for every 1 carboxyl group (per fatty acid molecule) with which to react, obviously an excess.

These known equations, however, represent only *theoretical* reactions (JA 12, fourth, third and second questions from the bottom). However, it has been clearly established by Plaintiffs-Appellants at Trial that *in practice*, it is impossible to obtain by the known reactions substantially pure diglyceride alone or substantially pure monoglyceride alone (JA 10, third question from bottom, JA 11, 12, 13, 17, 18 and 52a). Only the triglycerides can be obtained relatively pure when a stoichiometric amount of glycerine is used (JA 10, third question from the bottom, JA 52a).

The melting points of the pure triglycerides, diglycerides and monoglycerides of fatty acids having from 10 to 18 carbon atoms per molecule were also known per se prior to the invention of Plaintiff-

Appellant Baenitz (JA 52) and were readily available from standard textbooks (JA 10, first question and answer, referring to Plaintiffs' Exhibit 2).

However, the exact mechanism as regards the mixtures resulting from the esterification of fatty acids having from 10 to 18 carbon atoms per molecule, when varying the amounts of glycerine in excess of the stoichiometric amount, were totally unknown prior to the investigations and experiments attendant the discovery of Appellant Baenitz (JA 14), the knowledge of such reaction mechanisms being essential to an appreciation of the Baenitz invention and its correlation of melting point to hydroxyl number of the produced ester mixtures.

4. Evidence at Trial

At Trial, Plaintiffs-Appellants established without any doubt that pure diglycerides and pure monoglycerides could not be obtained *directly* by esterifying saturated fatty acids having from 10 to 18 carbon atoms per molecule with glycerine, regardless of the amount of excess glycerine used (JA 10, 11, 12, 13, 17, 18 and 52a).

Appellant Baenitz testified that in the direct esterification with glycerine, using the stoichiometric amount, approximately pure triglycerides could be obtained (JA 10, third question from bottom) but that substantially pure mono- and diglycerides could never be obtained regardless of the excess amount of glycerine used over and above the stoichiometric amount. Instead, either mixtures of tri- and diglycerides (JA 10, second question from bottom), or mixtures of tri-, di-, and monoglycerides (JA 10, first question from bottom) are obtained as also evidenced by Plaintiffs' Exhibit 3 (JA 52a), representing a graph showing the esterification products obtained when esterifying the same fatty acid with different amounts of excess glycerine (JA 11). The graph of Plaintiffs' Exhibit 3 (JA 52a) clearly establishes the fact that by using ever-increasing amounts of glycerine in excess of the stoichiometric amount, at first mixtures consisting primarily of triglyceride and small amounts of diglycerides are produced, with the amount of diglycerides increasing while the amount of triglyceride decreases, thereafter while the amounts of triglycerides further decrease and the amounts of diglycerides further increase, small amounts of monoglycerides are produced whereupon with an excess amount of glycerine producing an esterification product having a hydroxyl number about 150, small amounts of free glycerine are produced which increase with increase in the excess amount of glycerine used in the reaction, though the mixtures always contain substantial amounts of tri-, di- and monoglycerides (JA 11, 13). The results of the experiment of Plaintiffs' Exhibit 3 (JA 52a), indicating the various mixture compositions of, the resulting esterification product as a function of hydroxyl number are given in the tabulation above the graph of Plaintiffs' Exhibit 3 (JA 52a).

Additionally, the evidence based on known facts adduced at Trial in the form of Plaintiffs' Exhibit 2 (JA 52) also indicated that the higher the number of carbon atoms per molecule in the fatty acid, the higher the melting point. The same chart (Plaintiffs' Exhibit 2, JA 52), which was, as mentioned above, taken from standard textbooks also clearly establishes that the melting point of the diglyceride for a particular fatty acid is higher than the melting point for the triglyceride and that the melting point of the corresponding monoglyceride lies above the melting point of the corresponding diglyceride. These melting points as taken from the literature (JA 15) were also plotted on Plaintiffs' Exhibit 7 (JA 52d) in which the respective melting points indicated by square marks represent the melting points of the triglycerides (JA 15), the triangles represent the melting points for the corresponding diglycerides, and the circles represent the melting points for the corresponding monoglycerides. Plaintiffs' Ex. 7 (JA 52d),, like Plaintiffs' Ex. 4 (JA 52b), also indicates that within the claimed hydroxyl number range of 40 to 100, the curves for the melting points as a function of hydroxyl numbers initially extend downwardly approximately in the shape of a V or U, with the existence of a minimum of melting point within this range of hydroxyl numbers of 40-100, which lies considerably below the melting point of the corresponding triglyceride, the latter's melting point being the *lowest known* melting point for each particular glyceride. This minimum below the lowest previously known temperature as indicated in the literature is realizable for each of the fatty acids having about 10 to 18 carbon atoms per molecule and mixtures thereof as defined by the claims of the Baenitz application (JA 13 and 16). These graphs (JA 52b and 52d) were based exclusively on data found experimentally (JA 13) and indicate beyond any doubt that within the claimed range of hydroxyl numbers of 40 to 100 a *minimum of the melting point occurs which lies below the minimum known heretofore in the literature* (JA 13 and 16). Thus, it is an uncontroverted fact, established at trial, that for all the fatty acids falling within the claimed group, a melting point is obtainable by the use of the process of the Baenitz invention for the esterification products having a hydroxyl number of 40 to 100, which is lower than that of the corresponding pure tri-, di-, or monoesters (JA 14).

Mr. Hans-Herbert Schubert, head of the Patent Department of Plaintiff-Appellant Chemische Werke Witten, the assignee of the Baenitz application, testified that prior to the Baenitz discovery, it was customary and necessary to control the melting point by mixing various specially produced esters, indicating also that it was very difficult to mix these products to obtain a homogeneous product (JA 28).

Because of the further fact that the pharmaceutical ingredients, when added to any suppository masses may, in turn, somewhat raise or lower the melting point thereof, the pharmaceutical industry requires that the suppository masses not only be *hard and brittle* at ordinary temperatures but also be guaranteed to have a certain melting point (JA 28), such guarantee being a prerequisite to the successful sale of the suppository masses.

The testimony of Mr. Schubert also indicated that the desired and necessary control of the melting point is obtainable reliably and accurately for the first time by following the discovery of Mr. Baenitz and that the discovery disclosed in the Baenitz application enjoyed not only commercial success but permits Appellant Chemische Werke Witten to produce suppository masses with a definite melting point, unobtainable to date by its competitors (JA 29).

Aside from the importance of accurately controlling the melting point with the use of one and the same starting fatty acid (JA 9, next-to-last question), the lowering of the melting point, below the temperatures indicated for pure tri-, di- and monoglycerides, is also of significant importance as the fatty acids occurring in nature usually contain large amounts of fatty acids having from 10 to 18 carbon atoms, usually with the major portion of the fatty acids having 12 to 18 carbon atoms which would have a melting point far above that of the human body. Except for the triglyceride of capric acid, a C_{10} fatty acid, all tri-, di-, and monoglycerides of capric (C_{10}), lauric (C_{12}), myristic (C_{14}), palmitic (C_{16}), and stearic (C_{18}) acids have melting points from 44.5°C. (for the diglyceride of capric acid) and above, as known in the art (Plaintiffs' Exhibit 2, JA 52), which would render impossible their use as suppository bases since the normal body temperature is about 36°C.-37°C. so that they would not melt at body temperature. However, by being able to depress the melting point from that of the *lowest known* melting point for a given fatty acid ester, which is the melting point of the triglyceride of the respective fatty acid, the Baenitz invention permits the use of fatty acids as occur most frequently in nature in connection with the production of suppository masses notwithstanding the fact that the normally given and indicated melting points of their pure glycerides lie substantially above the human body temperature, there being no indication whatsoever in the prior art that the ester mixtures of the claimed hydroxyl range, produced by *direct esterification*, would have melting points lower than the lowest known melting point of any ester component therein.

STATUTES INVOLVED

35 U.S.C. 100. Definitions. When used in this title unless the context otherwise indicates—

> (a) The term "invention" means invention or discovery.
>
> (b) The term "process" means process, art or method, and includes a new use of a known process, machine, manufacture, composition of matter, or material.
>
> (c) The terms "United States" and "this country" mean the United States of America, its territories and possessions.
>
> (d) The word "patentee" includes not only the patentee to whom the patent was issued but also the successors in title to the patentee.

35 U.S.C. 101. Inventions Patentable. Whoever invents or discovers any new and useful process, machine, manufacture, or composition of matter, or any new and useful improvement thereof, may obtain a patent therefor, subject to the conditions and requirements of this title.

35 U.S.C. 102. Conditions for Patentability; Novelty and Loss of Right to Patent. A person shall be entitled to a patent unless —

> (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for patent, or
>
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or

on sale in this country, more than one year prior to the date of the application for patent in the United States, or

(c) he has abandoned the invention, or

(d) the invention was first patented or caused to be patented by the applicant or his legal representatives or assigns in a foreign country prior to the date of the application for patent in this country on an application filed more than twelve months before the filing of the application in the United States, or

(e) the invention was described in a patent granted on an application for patent by another filed in the United States before the invention thereof by the applicant for patent, or

(f) he did not himself invent the subject matter sought to be patented, or

(g) before the applicant's invention thereof the invention was made in this country by another who had not abandoned, suppressed, or concealed it. In determining priority of invention there shall be considered not only the respective dates of conception and reduction to practice of the invention, but also the reasonable diligence of one who was first to conceive and last to reduce to practice, from a time prior to conception by the other.

35 U.S.C. 103. Conditions for Patentability; Non-Obvious Subject Matter.

A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

STATEMENT OF POINTS

Appellants intend to rely on the following points:

1. The District Court erred in relying in its opinion, as material evidence, on documentary evidence neither offered nor received in evidence during the trial.

2. The Court erred in failing to grant Appellants' Motion to Vacate Judgment under Rule 59 F.R.C.P., relying instead, in its modified opinion, on evidence submitted by Appellee only after Trial and accepting arguments advanced by Appellee only after Trial which were based exclusively on this evidence submitted by Appellee after Trial.

3. Assuming the documentary evidence relied upon by the Court in its Order of January 28, 1965, had been properly introduced and received into evidence, it was nevertheless an error for the Court to assume that Christensen used 80% crude glycerine because (a) there is absolutely no evidence on which to base such assumption, (b) the intended use of the mono- and diglycerides of Christensen for edible materials and the use of the mixture compositions of tri-, di- and monoglycerides of Baenitz for suppository masses, particularly in the absence of evidence to the contrary, must be presumed conclusively to require the purest type of glycerine available, (c) in the absence of any evidence, the intended use as edible materials for the products of Christensen and as suppository masses for the products of Baenitz must be conclusively presumed to use the same type of glycerine, and (d) because any impurities in the glycerine would not only disturb the esterification of the stearic acid as proposed by Christensen but also the formation of the sodium glycerate catalyst disclosed by Christensen, which catalyst presupposes for its proper formation the presence of glycerine free of any impurities such as water.

4. The Court erred in holding that the processes of Christensen and of Plaintiff-Appellant Baenitz are essentially the same.

5. The Court erred in holding that the processes of Christensen and of Plaintiff-Appellant Baenitz would inherently produce the same results.

6. The Court erred in holding that the features of controlling the melting point would necessarily be included in the process of Christensen.

7. The Court erred in finding that the control of the melting point as claimed by Appellant Baenitz would have been obvious to a person skilled in the art as of the time the invention was made by Appellant Baenitz in view of the various textual publications and patents introduced by the Defendant at the Trial.

8. The Court erred in failing to properly apply Sections 102 and 103 of Title 35 U. S. Code as defined by Section 100, Title 35 U. S. Code, by failing to recognize patentability in the new use of a known process.

9. The Court erred in failing to recognize the criticality of the range of hydroxyl numbers as defined in the claimed process of Appellants' invention.

10. The Court erred in applying the "clearly erroneous" test in a trial de novo to the findings of the Patent Office.

11. The Court erred in failing to direct the Commissioner of Patents to allow the claims of Appellants' application.

SUMMARY OF ARGUMENT

1. In an action pursuant to U. S. C., Title 35, Section 145, it is improper for the Trial Court to predicate the rationale of its opinion on arguments advanced only and evidence submitted only after trial in the Brief for Appellee which evidence was never introduced nor received at trial; that the Trial Court relied on such arguments and evidence is borne out by the fact that in its Order of January 25, 1965, in

response to Plaintiffs-Appellants' Motion to Vacate Judgment under Rule 59 F.R.C.P., it assumed a use by Christensen of 80% crude glycerine, argued for the first time in Defendant's Brief after Trial at which time the Defendant-Appellee also submitted for the first time evidence for such newly presented arguments.

2. Since the entire evidence properly before the lower Court was devoid of any suggestion that Christensen used 80% crude glycerine, it was clearly erroneous for the lower Court to deny Plaintiffs-Appellants' Motion to Vacate Judgment under Rule 59, F.R.C.P.

3. Even if pages 636 and 637 of the publication "Industrial Oil and Fat Products" by Alton E. Bailey, the disputed evidence in question, which were attached to Defendant's Brief after Trial, had been properly introduced and received into evidence, and even if the arguments based thereon had been properly made at trial, it was nevertheless clearly erroneous on the part of the District Court to assume that Christensen used 80% crude glycerine and that Baenitz used 100% pure glycerine since there was absolutely no evidence whatsoever in the record to support such assumption; furthermore, in view of the intended use of the respective products, it was clearly erroneous for the Court to make such assumption only with respect to Christensen but not to apply the same reasoning to Baenitz. Additionally, the disputed evidence in question also indicated that for *commercial* purposes, the 80% crude glycerine has to be further purified; in view of the intended use of the product of Christensen as *edible* material, it was clearly erroneous for the District Court, in the absence of any evidence, to assume that Christensen would use 80% crude glycerine containing large amounts of harmful impurities. Additionally, the chemistry involved in the esterification of the fatty acids in question according to the Christensen process presupposes as pure a glycerine as possible as the presence of impurities would impair the proper chemical reaction necessary for such esterification.

4. Since Christensen uses an amount of glycerine (200-400 grams per 1,000 grams of fatty acid) for the production of his mono- and diglycerides which is substantially greater than the amount of glycerine used per 1,000 grams of fatty acid in the Baenitz process (131-168 grams), it was clearly erroneous for the lower Court to hold that the processes of Christensen and of Plaintiff-Appellant Baenitz are essentially the same.

5. Since the Christensen process produces only monoglycerides or diglycerides or mixtures of only mono- and diglycerides, and since the Baenitz process inherently produces mixtures of triglycerides, diglycerides and possibly small amounts of monoglycerides, it was clearly erroneous for the lower Court to hold that the process of Christensen would inherently produce the same results as that of Baenitz.

6. Since the products consisting of mono- and diglycerides and mixtures of mono- and diglycerides according to the Christensen process and the products of tri- and diglycerides with possibly small amounts of monoglycerides according to the Baenitz process are different from each other, it was clearly erroneous for the lower Court to hold that the particular control of the melting point within the claimed hydroxyl range would necessarily be included in the Christensen process, particularly as Christensen indicated no melting points whatsoever for his products and no hydroxyl numbers nor any critical range of hydroxyl numbers.

7. Admittedly, it was known in the prior art that the melting points of different tri-, di-, and monoglycerides are different from each other and that mixing such different glycerides would produce a mixture product which would have a melting point representing the average of the melting points of the mixture components. However, there is no indication or teaching, prior to the Baenitz invention, that within the claimed hydroxyl range of 40 to 100, the melting point of the claimed fatty acid esters could not only be accurately controlled

directly during the esterification of the fatty acid with glycerine but would inherently produce a *lowering* of the melting point *below* the lowest known temperature for the pure individual esters involved.

8. Though the esterification of fatty acids with glycerine, generally speaking, was known in the art, such general pre-existing knowledge, without the realization of the criticality of the particular hydroxyl range and the corresponding temperature control, i.e., lowering of melting points, does not bar the granting of a patent since the term "process" is defined in U. S. Code, Title 35, Section 100 as a "new use of a known process." Applying this statutory definition to the instant situation, it was clearly erroneous on the part of the lower Court to find that the control of the melting point as claimed by Appellant Baenitz would have been obvious to a person skilled in the art as of the time the invention was made by Appellant Baenitz.

9. In its opinion, the lower Court applied the "clearly erroneous" test to the findings of the Patent Office. Though great weight is normally given to the findings of the Patent Office, the proceedings in the trial Court are a trial de novo, and it was therefore clearly erroneous for the lower Court to apply the "clearly erroneous" standards of Rule 52(a) of the Federal Rules of Civil Procedure.

ARGUMENT

1. Reliance on Improper Evidence

Pursuant to Plaintiffs-Appellants' Motion to Vacate Judgment under Rule 59 F.R.C.P. (JA 35-41), the District Court modified its original opinion in an Order filed January 28, 1965 (JA 41). In the original opinion (JA 33), the District Court held:

> the amount of glycerine used in the Plaintiffs' process ranges from 131 to 168 grams per 1,000 grams of stearic acid, and that this clearly overlaps the Christensen range of 160 to 168 parts.

In the Motion to Vacate (JA 35 and 36), Plaintiffs-Appellants pointed out that the disclosure of Christensen prescribes the use of 200 to 400 grams of glycerine per 1,000 grams of stearic acid, and that therefore the overlap is, in fact, non-existent.

Since the entire rationale of the lower Court's opinion is based on this overlap as its major premise, as will appear more fully hereinafter, the Court modified its original opinion (JA 41), basing its conclusion on the *assumption*

> that 80% crude glycerine was used in the reaction (of Christensen).

However, there is no evidence whatsoever in the record at trial which would support the assumption of 80% crude glycerine. As a matter of fact, the Defendant-Appellee advanced no arguments during trial to the effect that Christensen should be assumed to have used 80% crude glycerine, Christensen being totally silent as to the type of glycerine used though it would have been easy at trial to demonstrate that Christensen, if anything, must be presumed to have used the purest type of glycerine since his mono- and diglycerides are intended as *edible* materials and since, from a chemical point of view, the existence of impurities in the glycerine would disturb, if not impair, the esterification reaction and production of the sodium glycerate catalyst of Christensen. The only suggestion of the existence of 80% crude glycerine was made in the Brief for Defendant after Trial in which, for the first time, it was argued that Christensen should be presumed to have used 80% crude glycerine. In support of this argument, the Defendant-Appellee attached to his Brief pages 636 and 637 of the "Industrial Oil and Fat Products" by Alton E. Bailey. However, this evidence was never introduced at trial, was not received into evidence and, therefore, was not properly before the Court in considering the question of patentability of the Baenitz invention.

Significantly, though the decision of the Board of Appeals of the U. S. Patent Office (JA 48-51) only relied upon three patents (JA 49), which were introduced as Defendant's Exhibit 1-A (JA 53 through 55), Defendant's Exhibit 1-B, (JA 56-59), and Defendant's Exhibit 1-C (JA 60-68), the Defendant-Appellee, apparently realizing the weakness of his position, introduced at trial the four additional Exhibits 2, 3, 4, and 5 (JA 69-77). Again, apparently after becoming aware of the continuing and fatal weakness of his position and to bolster the same, the Defendant attempted to introduce additional evidence after trial in his Brief after Trial.

It is basic that any document not offered and received in evidence is not in evidence and should not be relied upon as material evidence by the Court. *Kay et al. v. Cain*, 81 U.S.App.D.C. 24, 154 F.2d 205.

Obviously, the tactics of the Defendant deprived Plaintiffs-Appellants of an opportunity to rebut the evidence and to dispel any erroneous inference that might be drawn by the Court in the absence of such rebuttal, as was the case herein.

Furthermore, when arguments appear for the first time in the Briefs after Trial, the right of the affected party to introduce rebuttal evidence is basic to complete justice. *Douglas Aircraft Co. v. Mueller*, 107 U.S. App. D.C. 321, 277 F.2d 351.

Nor can there be any doubt that this evidence, improperly before the Court, was relied upon by the District Court as material evidence since the Court's own realization of the inaccuracy of its earlier opinion and the need for correcting the same exclusively on the basis of this evidence, indicates that without the assumption, even in the Court's mind, the original opinion would have been clearly erroneous.

It is therefore respectfully submitted that the reliance on this evidence, as material evidence, which was neither introduced nor received into evidence during the trial, which was not argued at any time during the trial and which was submitted to and urged upon the Court only after trial, was clearly erroneous.

2. Failure To Grant Motion To Vacate Judgment

In its original opinion, the lower Court found:

> The testimony demonstrated that the amount of glycerine used in the Plaintiffs' process ranges from 131 to 160 grams per 1,000 grams of stearic acid, and that this clearly overlaps the Christensen range of 160 to 168 parts. (JA 33).

In the Motion to Vacate Judgment under Rule 59 F.R.C.P. (JA 35-41), it was pointed out that the Christensen patent 2,022,494, on page 2, right column, lines 1 through 16 of the patent (JA 54), prescribes 120 parts by weight of glycerine for 300 parts by weight of stearic acid to produce a substantially pure monostearin.

On page 2, right column, lines 25 through 29 of this patent (JA 54), Christensen then teaches the preparation of substantially pure distearin by simply doubling the quantities of stearic acid used, i.e., by using 600 parts by weight of stearic acid for 120 parts by weight of glycerine. This means that Christensen teaches the use of

$$\frac{1{,}000}{300} \times 120 = 400 \text{ grams of glycerine}$$

to produce the disclosed monoglyceride with 1,000 grams of stearic acid, and

$$\frac{1{,}000}{600} \times 120 = 200 \text{ grams of glycerine}$$

to produce the disclosed diglyceride with 1,000 grams of stearic acid, as also pointed out in the Motion to Vacate Judgment (JA 35 and 36). Thus, having found

> that the amount of glycerine used in the Plaintiffs' process ranges from 131 to 168 grams per 1,000 grams of stearic acid, (JA 33)

it was clearly erroneous for the lower Court to hold that the range of 200 to 400 grams of glycerine per 1,000 grams of stearic acid overlaps the 131 to 168 grams of glycerine per 1,000 grams of stearic acid.

Since the remaining conclusions of the opinion are based on this overlap, and the overlap, in fact, does not exist, it was clearly erroneous for the lower Court to assume, in the absence of any evidence before the Court on which to base such assumption, a use by Christensen of 80% crude glycerine merely to re-establish the overlap.

Under these circumstances, it is respectfully submitted that it was clearly erroneous for the lower Court to deny Plaintiffs' Motion to Vacate Judgment under Rule 59 F.R.C.P. since the major premise of the Court's original decision, i.e., the alleged overlap in the amounts of glycerine used in the Baenitz process and in the Christensen process, would have been proper only if the assumption concerning the 80% crude glycerine had been proper.

3. Insufficiency of Additional Evidence

Assuming, *arguendo*, that pages 636 and 637 of the "Industrial Oil and Fat Products" by Alton E. Bailey had been properly introduced and received into evidence at trial, it was, nevertheless, clearly erroneous for the lower Court to assume that Christensen used 80% crude glycerine and Baenitz used 100% pure glycerine. Pages 636 and 637 of this publication are attached to this Brief as Appendix 1.

These two pages merely describe the recovery of glycerol from spent lyes which constitutes an important operation in any soap plant, as glycerol is an important by-product of soap making.

The publication points out that spent lyes contain, in addition to about 5% to 15% glycerol, *a considerable and variable quantity of salt, a small amount of free caustic soda, traces of dissolved soap and certain organic impurities derived from the fat.*

In the course of the recovery, the lye is treated to obtain at first "half-crude" glycerine, i.e., a solution with a glycerol concentration of about 40%. Thereafter, the half-crude glycerine is concentrated to

80% crude glycerine which still contains "*a considerable amount of salt and various other impurities, both organic and inorganic.*" The 80% crude glycerine is then purified and further concentrated

> to yield the *commercial* grades of C.P. glycerine, U.S.P. glycerine, dynamite glycerine, etc., . . . (emphasis added)

Since the monoglycerides and diglycerides produced by Christensen are intended for use with edible materials, as described on page 1, left column, lines 6 through 15 of the patent (JA 53) and on page 3, left column, lines 2 and 3 of the patent (JA 55), the assumption that Christensen used 80% crude glycerine containing "a considerable amount of salt, and various other impurities, both organic and inorganic" is totally unwarranted, particularly as page 636 clearly points out that the *commercial grades* are those which result from further purification of the 80% crude glycerine by stating:

> Purification and further concentration of this product (80% crude glycerine), to yield the *commercial* grades of C.P. glycerine, U.S.P. glycerine, dynamite glycerine, etc., is carried out by fractionally distilling the glycerol. (emphasis added).

In that connection, the attention of this Court is also respectfully directed to page 2, right column, lines 53 through 56 of the Christensen patent (JA 54), where the patentee stated:

> This is because the mono- and diglycerides are generally incorporated in very small amounts in *edible* oils, etc., and the quantity of the catalyst, for example, is so small as to be quite *harmless*. (emphasis added)

This concern by Christensen for purity and possible harm in the edible materials using the Christensen products clearly precludes the assumption that Christensen intended to use 80% crude glycerine in view of the numerous and harmful impurities that still exist in the 80% crude glycerine.

Additionally, since Baenitz and Christensen disclose the use of their ester products in edible materials and pharmaceutical products, and since there is no evidence whatsoever in the record as to the type of glycerine used by either, it was clearly erroneous for the Court, in the absence of any suggestion in the evidence, to assume Christensen used 80% crude glycerine and Baenitz used 100% pure glycerine. In view of the similarity of the final use of the products of Christensen and Baenitz, and in the absence of any clear evidence of the use by Christensen and Baenitz of different types of glycerine, the assumption made by the Court to correct the technical inaccuracy of its initial opinion was clearly erroneous.

Furthermore, it could have been readily established at trial by appropriate rebuttal evidence that the chemical reactions for the esterification of higher fatty acids with glycerine, producing esters and water, as indicated above, would be impaired by the presence of impurities containing water, normally present in 80% crude glycerine, since it is well known to any chemist that the presence of one of the end products resulting from a chemical reaction impairs such reaction as it has a tendency to bar or even reverse the reaction.

Under these circumstances, even if the pages in question had been properly received in evidence, it would have been clearly erroneous for the lower Court to assume that Christensen used 80% crude glycerine and Baenitz used 100% pure glycerine, the only possible explanation for such an assumption being the apparent desire on the part of the lower Court to re-establish the overlap of its original decision, without which the entire original opinion would be void.

Additionally, Plaintiffs would have been able to clearly rebut at trial the assumption of the use of 80% crude glycerine by evidence obtained from Defendant's own Exhibit 1-B, the Feuge et al patent, U.S. 2,745,749. The Feuge et al patent, like the Christensen patent, is mainly concerned with *edible* products (Col. 1, lines 41-43; Col. 1, line 71; Col. 2, lines 1-3; Col. 2, lines 59-64; and Col. 4, lines 51-52). Feuge et al. describe the production of three products in their patent — monostearate A, monostearate B, and monostearate C (Col. 2, line 70).

Monostearate A was produced by the method of Gros and Feuge, *J. Am. Oil Chemists' Soc.*, Vol. 28, pp. 1-4 (1951) (Feuge et al patent, Col. 2, line 70 to Col. 3, line 1). Page 1 of the Gros and Feuge article is attached to this Brief as Appendix 2. The method thereof is set forth at the bottom of the first column of page 1 of the article under "Materials and Methods." Herein it is stated:

> The fatty acids and methyl esters were converted to the corresponding monoglycerides by allowing them to react with *U.S.P. glycerol* . . . (emphasis added).

As stated above, U.S.P. glycerol (or glycerine) is substantially 100% pure glycerine.

Monostearate B of Feuge et al was a commercial, molecularly distilled product (Col. 3, lines 8-9). Being molecularly distilled, this product is obviously pure, and, as pointed out above with respect to pages 636 and 637 of the Bailey citation, the *commercial* grades of glycerine are C.P., U.S.P. or dynamite — all substantially 100% pure.

Monostearate C of Feuge et al was a technical grade product made from completely hydrogenated cottonseed oil (Col. 3, lines 13-15). Such technical grade products are referred to by Gros and Feuge (Appendix 2 attached hereto) in the second paragraph of column 1 on page 1 thereof, citing the paper by Feuge, *J.Am. Oil Chemists' Soc.*, Vol. 24, pp. 49-52 (1947). Page 49 of this latter paper by Feuge is attached to this Brief as Appendix 3. Feuge discusses therein mono- and diglycerides used in making products such as shortenings and margarine (Col. 1, line 4). The preparation of such technical grade mono- and diglycerides is set forth in the paragraph under "Experimental" in column 1 of the 1947 Feuge paper. Herein it is stated that the mono- and diglycerides therein are prepared by reacting hydrogenated cottonseed oil with "varying percentages of *U.S.P. glycerol*" (emphasis added), again substantially pure 100% glycerine.

Thus, it is abundantly clear that *substantially pure 100% glycerine, not 80% crude glycerine*, is used in preparing products to be employed in food or pharmaceutical uses. To *assume* that Christensen used *80% crude glycerine* to make *edible* products merely to create an overlap with Plaintiffs' claimed process is indeed clearly unwarranted.

4. Identity of Christensen and Baenitz Processes and Products

Since Christensen uses 200 to 400 grams of glycerine for 1,000 grams of stearic acid to produce the mono- and diglycerides, which is substantially greater than the amount of 131 to 168 grams of glycerine per 1,000 grams of stearic acid used by Baenitz, ignoring the clearly erroneous assumption of 80% crude glycerine by Christensen only, the Court's finding

> From this, it appears that the processes of Christensen and the Plaintiffs are essentially the same. (JA 33)

is believed clearly erroneous.

As pointed out above, the teachings of 200 to 400 grams of glycerine per 1,000 grams of stearic acid by Christensen are taken from the latter's disclosure on page 2, right column, lines 1 through 29 (JA 54) while the use in the Plaintiffs' process from 131 to 168 grams per 1,000 grams of stearic acid was also the finding of the Court in the original opinion (JA 33).

With respect to Plaintiffs' Exhibit 6 (JA 52c), Appellant Baenitz testified that the graphs represent calculated amounts of glycerine necessary to react with 1,000 grams of fatty acid to obtain the monoglycerides, shown by the blue line, to obtain the diglycerides, shown by the green line, and the triglycerides, shown by the red line (JA 14, last four questions and JA 15, first question). With respect to the black lines of Plaintiffs' Exhibit 6 (JA 52c), the witness Baenitz testified that they represent the glycerine requirement for esters with hydroxyl numbers within the claimed range of 40 to 100, the center lines indicating the preferred range of hydroxyl numbers of 60 to 80 (JA 15, second and third question.)

Interpreting Plaintiffs' Exhibit 6 in relation to the Christensen patent, Dr. Morrell, who was duly qualified as an expert witness, pointed out that the mono- and diglycerides according to the teachings of Christensen would fall between the blue and green lines of Plaintiffs' Exhibit 6 (JA 20, last question and JA 21, first question and JA 22, first question).

Admittedly, pure diglycerides have a hydroxyl number greater than 80 and less than 100 when produced from fatty acids having 16 and 18 carbon atoms per molecule. For example, the diglycerides of palmitic acid (C_{16}) has a hydroxyl number of 98.6 (the corresponding triglyceride having a hydroxyl number of 0 and the corresponding monoglyceride having a hydroxyl number of 339.6) while the diglyceride of stearic acid (C_{18}) has a hydroxyl number of 89.8 (the corresponding triglyceride again having a hydroxyl number of 0 and the corresponding monoglyceride having a hydroxyl number of 313), as evidenced by Plaintiffs' Exhibit 2 (JA 52). Much emphasis has been placed by the Defendant on this overlap in the hydroxyl number between *pure* diglyceride and the claimed range of hydroxyl numbers of 40 to 100 of ester *mixtures* produced by the direct esterification of fatty acids having from 10 to 18 carbon atoms per molecule. However, whereas the overlap with the claimed hydroxyl range of 40 to 100 exists between the claimed esters of the Baenitz process and certain *pure diglycerides*, it is an uncontroverted fact that the esterification of the claimed fatty acids with an excess amount of glycerine to produce the claimed hydroxyl range of 40 to 100 according to the Baenitz process results in ester mixtures consisting primarily of triglycerides and diglycerides with possibly small amounts of monoglycerides. In that connection, reference is made to the testimony of the inventor Baenitz who testified that the esterification of a fatty acid with an excess amount of glycerine inherently produces tri- and diglycerides or tri-, di-, and monoglycerides (JA 10, last two questions).

That the ester mixtures produced according to the Baenitz process from the esterification of fatty acids with the claimed excess glycerine to produce a hydroxyl number of 40 to 100 is different from *pure* diglyceride was also clearly established at trial (JA 17). On re-direct examination, and to point out the difference within the area of overlap of Plaintiffs' Exhibit 6 between the pure diglicerides and the products obtained by the Baenitz process, the inventor Baenitz testified as follows:

> Q. Let me rephrase the question. If you use stearic acid with C_{18} and you use an excess quantity of glycerol to obtain a hydroxyl number of 90, what would you obtain in the mixture?
>
> A. Tri- and diglyceride.
>
> Q. Would you get a small amount of mono-?
>
> A. And a small amount of mono- under 10%.
> (JA 17, third and second question from the bottom)

The difference between the *pure diglyceride* and the *mixture* products obtained by direct esterification of the claimed fatty acids according to the Baenitz process was also clearly established by the following testimony (JA 18, first and second questions).

The ester mixtures obtained by direct esterification of the same fatty acids with different amounts of excess glycerine in accordance with the Baenitz method are shown in Plaintiffs' Exhibit 3 (JA 52a) which clearly establishes that within the claimed range of hydroxyl numbers of 40 to 100, the ester mixtures obtained by the Baenitz process would consist primarily of tri- and diglycerides with small amounts of monoglycerides at the upper end of this range.

Since Christensen teaches the production by esterification of only monoglycerides, diglycerides, or mixtures of mono- and diglycerides (JA 20, 21 and 26) and since the ester mixtures produced by the Baenitz process consist of triglycerides, diglycerides and small amounts of monoglycerides (JA 52a), it was clearly erroneous for the Court to find:

> Accordingly, it seems reasonable to suppose they (Christensen and Baenitz) would inherently produce the same results. (JA 33).

The next sentence of the Court is even more astounding in the light of the clear and convincing evidence adduced at trial:

> At least, the Court cannot say such a view is unwarranted where Plaintiffs have offered no convincing proof to the contrary. (JA 33).

All the evidence at trial as well as the Christensen patent itself (JA 53 to 55) indicates that Christensen contemplated the production of only monoglycerides and diglycerides or mixtures of mono- and diglycerides whereas the Baenitz process inherently produces mixtures of tri- and diglycerides with the possibility of small amounts of monoglycerides. Thus, the evidence clearly established that the alleged product obtained by the Christensen process is very different from that obtained by the Baenitz process.

Since the processes of Christensen and Baenitz are different as pointed out above, and since the products inherently produced by the respective processes of Christensen and of Baenitz are different, as also pointed out above, the Court's syllogism of

> On the question of controlling the melting point, it is clear, first that if Christensen's process inherently produces Plaintiffs' result, then the feature of controlling the melting point would necessarily be included in such process (JA 33)

was also clearly erroneous. It is also significant in that connection that Christensen nowhere mentions the hydroxyl number or a melting point of the produced glycerides. Nor is there any finding in the decision that Christensen taught the control of the melting point within the claimed range of hydroxyl numbers.

5. Obviousness of Invention

On the question of controlling the melting point, the Court found:

> Whether this advantage in fact occurred to Christensen appears somewhat doubtful. It is clear, however, from the other references, that varying the proportion of ingredients in mixtures such as Christensen's was, at the time of the invention, known to vary the mixture's melting point. This was shown by the various textual publications introduced by the Defendant at trial, and also by the patent to Feuge et al. (JA 33 and 34).

Admittedly, it was known in the prior art, as already indicated in Plaintiffs' Exhibit 2 (JA 52), that the pure esters of the various fatty acids having 10 to 18 carbon atoms, which have different hydroxyl numbers, also have different melting points. Plaintiffs' Exhibit 2 (JA 52) which was prepared from standard textbooks, indicates in the column "Smp." the melting points for the various *pure* triglycerides, diglycerides, and monoglycerides. Hence, the mixing of various pure esters would also be expected to produce mixtures having melting points different from those of the individual pure constituent components. As a matter of fact, this very mixing was also used prior to the Baenitz invention by Appellant, Chemische Werke Witten, the assignee of the Baenitz application, to obtain suppository masses with the desired melting points (JA 28). However, the Baenitz invention is not concerned with controlling the melting point by *mixing* the various ingredients, but is concerned with *directly* controlling the melting point *during the esterification* as is also abundantly clear from claim 25, a representative claim of the Baenitz application, which calls for:

> A method for controlling the melting point of *hard and brittle fats* within a relatively *narrow desired* range *during the manufacture thereof by esterification* of a fatty acid . . . with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, . . . (JA 30) (emphasis added).

In other words, it is not by mixing various components that the Baenitz invention seeks to control the melting point but by the chemical reaction directly during the esterification with a certain excess amount of glycerine to produce the claimed range of hydroxyl numbers of 40 to 100. Thus, the Baenitz discovery is neither concerned with controlling the melting point by mixing various ingredients nor with generally varying the melting point, but is limited to the method of controlling the melting point directly during the esterification, i.e., during the chemical reaction of the fatty acid with glycerine, with such an excess amount of glycerine (in excess of the stoichiometric amount) as to produce a hydroxyl number of 40 to 100 in the ester product, inherently causing the aforementioned lowering of the melting points.

Referring to Plaintiffs' Exhibits 4 and 7 (JA 52b and 52d), representing graphs for the melting point of fatty acid esters in dependence upon the hydroxyl number, experimentally obtained by Appellant Baenitz (JA 13, 15), it is clear that for each fatty acid having between 10 and 18 carbon atoms per molecule (Plaintiffs' Exhibit 7, JA 52d), and for each ester mixture produced from mixtures of various fatty acids having 10 to 18 carbon atoms per molecule (Plaintiffs' Exhibit 4, JA

52b), there exists a minimum melting point which is lower than the lowest known melting point indicated in the standard prior art textbooks (JA 52). Turning to Plaintiffs' Exhibit 7 (JA 52d), the curves representing the melting point as a function of hydroxyl number are all of approximately V shape, with the melting point decreasing from the zero hydroxyl number corresponding always to a given triglyceride, to a given minimum within the range of hydroxyl numbers of 40 to 100. This minimum is clearly below that of the melting points indicated in the literature for the pure triglyceride (JA 52 and 52d), which has the lowest known melting point for a pure glyceride produced from a given fatty acid. Hence, the question presented by the present case is instead "whether it was obvious to control the melting point during the esterification of the claimed fatty acids with such an excess amount of glycerine as to obtain in the inherently resulting ester mixtures a hydroxyl number falling within the claimed range of 40 to 100, *whereby the melting temperature within this range is inherently lowered* from the lowest known melting point of the constituent components of the corresponding pure esters." Christensen never mentions melting points nor hydroxyl numbers (JA 20) and even the Court below doubted that this advantage in controlling the melting point "occurred to Christensen." (JA 33).

The Feuge et al patent (JA 56-59) uses as starting material the long chain saturated fatty acids that may be the final product of the Baenitz process (JA 33). As a matter of fact, the Court held that the Feuge et al patent:

> describes a process for modifying the melting point and like properties of *monostearin* by directly acylating a portion of the hydroxyl groups of monostearin. (JA 31) (emphasis added).

However, monostearin is an ester produced by the reaction of stearic acid (C_{18}) with glycerine and constitutes one of the reaction products of Christensen as well as one of the components, though in small proportions, of the ester mixtures obtained by the Baenitz process when using stearic acid and glycerine. Thus, the Feuge et al patent is concerned with changing the characteristics of an end product as obtained by the Baenitz process.

The *"acylated"* products obtained by Feuge et al are flexible and stretchable (JA 56, column 2, lines 25 to 27), whereas suppository masses must be *hard and brittle*, as claimed. The hydroxyl value of the acylated mixture indicated by Feuge et al is *"between about 15 to 200"* (JA 56, column 1, lines 68 to 70), and no criticality is indicated as to the melting points for the claimed hydroxyl range of 40 to 100. As a matter of fact, Tables II and III (JA 58) of this patent merely indicate, what is admittedly already known; namely, that the melting point, as expected, increases with increase in the hydroxyl number, as borne out also by Plaintiffs' Exhibit 2 (JA 52) and by Plaintiffs' Exhibit 7 (JA 52d). However, it is respectfully submitted that this prior art knowledge indicates no criticality within the claimed range of hydroxyl numbers of 40 to 100 for the claimed fatty acid esters nor does such information indicate the possibility of *depressing* the melting point of the claimed fatty acid ester mixtures within this claimed hydroxyl range, as is possible by the Baenitz method. The Feuge et al patent certainly does not teach nor even suggest the criticality of the claimed hydroxyl range nor the fact that within this particular critical range a particular lowering of the melting point of a fatty acid ester occurs as claimed by Appellant Baenitz; all that Feuge et al. teach is that *acylated monoglycerides* can be produced whose melting points *increase* with an increase of the hydroxyl number, as would be expected from the literature (JA 52 and 52d), compared to a *decrease* of the melting points for the Baenitz products with increase of the hydroxyl number in the claimed range.

Turning to the other evidence introduced at trial by the Defendant, Defendant's Exhibit 2 (JA 69, 70) merely confirms the information of

Plaintiffs' Exhibit 2 (JA 52) insofar as the characteristics of glycerol monostearate are concerned. Defendant's Exhibit 3 (JA 71-73) merely dealt with the question of determination of the purity of a product, stating:

> As the presence of some other substance lowers the melting point *in proportion* to the quantity of it present, an accurate determination of melting point is a ready means of establishing the purity of a product. (emphasis added).

Since the melting point of the "other substance" in relation to that of the product is not indicated, and since this aspect only deals with the question of *purity* of a product contrasted with melting point of a *reaction mixture,* certainly this publication constitutes no teaching for the criticality within the claimed hydroxyl range of 40-100 nor for the novel, unexpected results in the lowering of the melting point of the fatty acid mixtures.

Defendant's Exhibit 4 (JA 74, 75) states under 4. Calculation of Ideal Solubility:

> It is well known that the melting point of a substance is usually lowered by the *admixture* of a small proportion of any second substance, even if the latter is relatively high melting. (emphasis added).

This paragraph, which deals with "Calculation of Ideal Solubility," merely points out possible variations by the *"admixture"* of a *"small"* proportion of a second substance which is different from a *chemical reaction* producing a *mixture* essentially consisting of substantial proportions of two ingredients; namely, of tri- and diglycerides, as is the case with the Baenitz method. This was also clearly understood by the Court when it queried the witness:

> THE COURT: Well, of course, when you have an admixture you have no chemical reaction do you? (JA 27)

Under these circumstances, since there is no evidence whatsoever in the record to suggest that within the *claimed hydroxyl range* the melting point of the *claimed fatty acid esters* could be lowered below the lowest melting point known for any of its pure components, it is respectfully submitted that the Court's holding as to obviousness of the Baenitz process was clearly erroneous.

The importance of being able to lower the melting point of the ester mixtures produced from fatty acids having 10-18 carbon atoms was also clearly brought out at trial (JA 9, 10) since the Baenitz invention permits the use of fatty acid mixtures as occur in nature for the use in the production of suppository masses which, without the benefit of the Baenitz invention, would have excessively high melting points above the body temperature.

Section 100 of Title 35 of the U.S. Code clearly recognizes patentability for a discovery even in the "new use of an old process," a definition applicable to the instant situation if it were argued that the esterification of fatty acids, generally speaking, was known before the Baenitz discovery.

6. Burden of Proof

In its decision, the lower Court applied the "clearly erroneous" test in holding:

> In view of this, the Court cannot say the Patent Office was *clearly wrong* in finding that the control of the melting point as claimed by Plaintiffs, would have been obvious to a person skilled in the art having all these disclosures before him. (JA 34) (emphasis added).

While it has been repeatedly held to be proper for the trial judge to place great weight on the findings of the Patent Office, *Esso Standard Oil Co. v. Sun Oil Co.*, 97 App. D.C. 154, 229 F.2d 37; *Zenith Corporation v. Ladd*, 114 App. D.C. 54, 310 F.2d 859, it is nevertheless true that the trial before the District Court was a trial de novo. *Johns-Manville Corporation v. David L. Ladd*, 117 App. D.C. 262, 328 F.2d 563. To apply in the District Court the "clearly erroneous" test as it is used in this Court under Rule 52(a), F.R.C.P. in itself was a clearly reversible error of the lower Court.

CONCLUSION

For the reasons given above, Plaintiffs-Appellants respectfully submit that the lower Court erred in ordering judgment for the Defendant, in denying Plaintiffs-Appellants' Motion to Vacate Judgment and in failing to find for Plaintiffs-Appellants.

Respectfully submitted,

PAUL M. CRAIG, JR.

DICKE & CRAIG
Pennsylvania Building
Washington, D. C.

Attorney for Appellants

In the above process, a factor to be considered is the possible loss of glycerol through decomposition at the high operating temperatures employed. The rate of destruction of glycerol in contact with alkalies at high temperatures is the subject of a recent communication by Mattikow and Cohen.[11]

(e) *Recovery of Glycerol*

The recovery of glycerol from spent lyes is an important operation in any soap plant, as glycerol is an important by-product of soapmaking.

The spent lyes contain, in addition to about 5–15% glycerol, a considerable and variable quantity of salt, a small amount of free caustic soda, traces of dissolved soap, and certain organic impurities derived from the fat. The first step in glycerol recovery is treatment of the lye with aluminum sulfate and sulfuric or hydrochloric acid. The addition of about 4 to 14 pounds of aluminum sulfate to each 1000 pounds of lye precipitates any dissolved soap in the form of insoluble aluminum salts, and reacts with the excess caustic soda to form a flocculent precipitate of aluminum hydroxide which has a high adsorptive capacity for albuminous material or other organic impurities. Sufficient acid is also added to neutralize any caustic soda remaining after reaction with the aluminum sulfate. After treatment is completed the clarified lye is filtered in ordinary filter presses.

The treated lye is evaporated to precipitate the bulk of the salt, and concentrate the solution to so-called 80% crude glycerine. Vertical-tube, double effect evaporators are commonly used for evaporating the solution to a glycerol concentration of about 40%, at which point it is known as "half-crude" glycerine. Separation of the bulk of the salt occurs in the second effect, the salt settling to the bottom of the evaporator and thence falling into salt drums. Valves are placed in the pipes leading from the evaporators to the salt drums, so that the latter may be periodically cut off from the evaporator proper, and emptied. The recovered salt, after washing and drying, is reused for salting out new batches of soap.

In the further concentration of half-crude to 80% crude glycerine, the first effect is cut out of the system, or a separate single effect evaporator is used, high-pressure steam being employed in either case for heating the solution. With the usual vacuum of 26 to 28 inches on the evaporator, a concentration of 80% will be reached when the temperature has risen to about 190° to 200°F.; excessive losses of glycerine will occur if further concentration is attempted.

Ordinary 80% crude glycerine contains a considerable amount of salt, and various other impurities, both organic and inorganic. Purification and further concentration of this product, to yield the commercial grades of C.P. glycerine, U.S.P. glycerine, dynamite glycerine, etc., is carried out by fractionally distilling the glycerol from the nonvolatile impurities under vacuum, with the aid of superheated steam, and treating the distilled product with bleaching carbon and various chemicals.

---

[11] M. Mattikow and B. Cohen, *Oil & Soap*, 20, 135–136 (1943).

2. Solidification and Drying of the Liquid Soap

(a) *Framing and Drying of Framed Soaps*

The simplest method of converting neat soap, or other hot liquid soap to a solid form suitable for forming into bars, is known as framing. This consists simply of running the liquid soap into frames and allowing it to solidify spontaneously in the form of large cakes. Various coolers have been devised in which the soap is quickly solidified in small, water cooled frames, but these do not appear to have attained wide use, at least in the United States.

The frames consist of a bottom, mounted on wheels, and four removable sides, which can be clamped together to form a liquidtight compartment, and yet be readily removed from the solidified cake. A frame of soap usually weighs 1000 to 1200 pounds, and is shaped in the form of a high, narrow rectangle, for maximum convenience and economy in subsequent cutting operations. About 3 to 5 days are ordinarily required for the solidification of neat soap; cold made soaps may require slightly longer, since saponification is completed in the frames.

The frames of soap are cut into slabs and then into cakes by being forced through metal frames set with thin, taut wires. The cakes or blanks resulting from this operation must be dried to some extent before they can be stamped and wrapped. Drying is carried out by passing the blanks through drying rooms or tunnels, where they are subjected to the action of warm, dry, circulating air. Drying occurs largely on the surface, hence the moisture content of the whole bar is not usually reduced by more than 3% to 5%.

Floating soaps produced by framing simply contain a considerable proportion of air beaten into the soap mass in the crutcher. This air is in the form of very fine, almost invisible bubbles, which are readily retained by the viscous soap entering the frames.

Added materials, such as builders, antioxidants or stabilizers, perfumes, etc., are crutched into the kettle soap before framing.

(b) *Manufacture of Soap Chips*

Soap chips are manufactured not only to be packaged as such, but also to be milled and plodded into toilet soap bars, the solidification and drying of soap in the form of chips having in modern plants entirely supplanted other methods of preparing neat soap for milling.

Solidification of soap for the production of chips is accomplished on a

LIBRARY
JAN 16 1951
U.S. PATENT OFFICE

The Journal of the American Oil Chemists' Society

Volume 28 JANUARY, 1951 No. 1

Modification of Vegetable Oils. X. Effect of Monoglycerides on the Interfacial Tension of Oil-Water Systems[1]

AUDREY T. GROS and R. O. FEUGE, Southern Regional Research Laboratory,[2]
New Orleans 19, Louisiana

MONOGLYCERIDES are employed widely as surface active agents in the preparation of oil-water emulsions. One of the early uses of these compounds for such a purpose was in the manufacture of "superglycerinated" or "high ratio" shortenings, and today this application is still considered to be the most important. However monoglycerides are used in substantial quantities in other types of emulsions, especially in the manufacture of cosmetic and pharmaceutical preparations. Cold creams, for example, may consist almost entirely of technical grade monostearin, mineral oils, and water.

Despite the widespread use of monoglycerides as emulsifiers few quantitative data have been published concerning their performance in this field. In a previous article (2) the effectiveness of technical grade monoglycerides (prepared from cottonseed oil stearine) in lowering the interfacial tension at vegetable oil-water interfaces was reported. Subsequently Kuhrt *et al.* (5) reported data on the interfacial tension of a number of monoglyceride-vegetable oil-water systems.

The present investigation was undertaken to obtain additional data on the behavior of monoglycerides in oil-water mixtures, particularly the monoglycerides monopalmitin, monostearin, mono-olein, and monolinolein. The monoglycerides of caprylic, capric, lauric, and myristic acids also were prepared and tested for comparative purposes.

Materials and Methods

Monoglycerides. The fatty acids employed in the preparation of monostearin and monopalmitin were prepared from commercial products (90% purity) by repeated fractional crystallization from acetone. The other saturated acids used in this investigation were obtained in relatively pure form from a chemical supply house. Each product contained 98% or more of the desired acid. Mono-olein was prepared from methyl oleate (approximately 97%) isolated from the mixed methyl esters of pecan oil fatty acids by repeated fractional crystallization from acetone. Monolinolein was prepared from methyl linoleate (approximately 95%) obtained from the mixed methyl esters of cottonseed oil by a similar fractionation procedure.

The fatty acids and methyl esters were converted to the corresponding monoglycerides by allowing them to react with U.S.P. glycerol for 3 hours at 200°C. in the presence of 0.1% of sodium hydroxide (on a fatty acid or ester basis) as catalyst.

In all cases the reaction was carried out under hydrogen, and the maximum amount of glycerol judged to be reactable and miscible with the fatty phase under these conditions was used in each case. Soap and unreacted glycerol were removed from the reaction products by acidifying with a 1% solution of acetic acid and washing with distilled water. In the case of the monoglycerides of the unsaturated acids a small amount of ethyl acetate also was added to facilitate washing. The products then were dried by melting them under vacuum and stripping with hydrogen.

The monoglycerides of the saturated fatty acids and mono-olein were concentrated by fractional crystallization from solvents. A solution containing 70% isopropanol was used for crystallizing monostearin and monopalmitin, and commercial hexane was used for the others. Residual solvent was removed from the concentrated products by melting under vacuum and stripping with hydrogen.

The products were analyzed for monoglyceride content by the periodic acid oxidation method as modified by Handschumacher and Linteris (3). The following values were obtained: monocaprylin, 92.8%; monocaprin, 96.2%; monolaurin, 86.9%; monomyristin, 78.6%; monopalmitin, 74.8%; monostearin, 86.2%; mono-olein, 88.6%; and monolinolein, 53.9%.

The products could have been further crystallized to increase their purity, but the procedures are time-

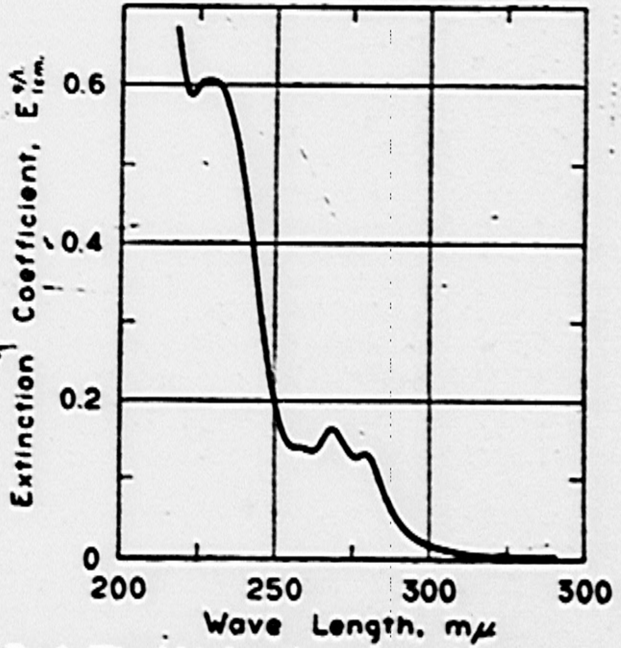


FIG. 1. Ultraviolet absorption spectrum of monolinolein

---

[1] Presented at the 41st Annual Meeting of the American Oil Chemists' Society, Atlanta, Ga., May 1-3, 1950.

[2] One of the laboratories of the Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.

77736

APPENDIX 3

Interfacial Tension of Oil-Water Systems Containing Technical Mono- and Diglycerides[1]

R. O. FEUGE
Southern Regional Research Laboratory[2]
New Orleans, Louisiana

TECHNICAL mono- and diglycerides find wide industrial use in the manufacture of products such as oil-modified alkyd resins, superglycerinated shortenings, and margarine. Probably the major uses of mono- and diglyceride preparations depend upon the ability of these substances to act as oil-soluble emulsifying agents.

It is generally recognized that monoglycerides are more active as surface-modifying agents than are the diglycerides; at present however, it is very difficult, if not impossible, to prepare, by a commercial process, a product consisting substantially of monoglycerides. Commercially available products are composed of a mixture of mono-, di-, and triglycerides. Acceptance of products intended for use as emulsifiers is generally dependent on the interfacial tension of fatty solutions of the products against water.

The literature contains little data on the interfacial tension of systems containing monoglycerides or mixtures of mono- and diglycerides. The scarcity of such data probably results from the fact that until recently no reliable method was available for determining the proportions of mono-, di-, and triglycerides in mixtures of these compounds.

The interfacial tension measurements reported here are intended to supply some quantitative data relative to the effectiveness of technical monoglycerides and diglycerides in lowering the interfacial tension of water against vegetable oils.

Experimental

The technical mono- and diglycerides used in these experiments were prepared from the same materials and by the same method described by Feuge and Bailey (3). Briefly the method is as follows: refined and bleached cottonseed oil, hydrogenated to an iodine value of 0.7, was reacted with varying percentages of U.S.P. glycerol while being stirred in a stainless steel-glass apparatus and maintained under hydrogen at one atmosphere pressure. Sodium hydroxide equal to 0.1% of the weight of fat was used as catalyst, and the reaction was continued for 5 hours at 200° C. (unless otherwise noted). The mean molecular weight of the fatty acids obtainable by saponification of the interesterified products was 279.4, calculated from the neutralization value of the acids.

The free glycerol and the soaps present in the reaction mixtures were removed. Free glycerol was eliminated by washing with 20% sodium sulfate solution. Removal of all traces of soap necessitated acidifying the mixtures. Under the conditions employed dilute mineral acids could not be used to destroy soap because these acids tended to decompose the monoglycerides. Repeated washing with a 20% sodium sulfate solution containing monobasic sodium orthophosphate and dibasic sodium orthophosphate in a molar concentration of 0.1 and 0.025, respectively, was found to decompose the soaps without affecting the monoglycerides. Decomposition of the soaps was followed by several washings with 20% sodium sulfate solution after which the products were dried and filtered.

The purified products were analyzed for total combined glycerol by the bichromate method of the American Oil Chemists' Society (1), after alcoholic saponification and removal of the alcohol by the procedure of Smith and Matthews (6). They were also analyzed for monoglyceride content by the periodic acid oxidation method of Pohle, Mehlenbacher, and Cook (5). From the combined glycerol, monoglyceride, and free fatty acid contents, and the mean molecular weight of the fatty acids, the compositions of the different products were calculated in terms of mono-, di-, and triglycerides. The results of the calculations of the glyceride distribution are given in Table 1.

TABLE 1

Composition of Monoglyceride-Diglyceride Products

Product	Composition,[a] weight percent		
	Monoglyceride	Diglyceride	Triglyceride
1	1.40	17.57	80.38
2	10.25	41.06	48.04
3	17.92	45.49	35.94
4	27.01	48.32	24.02
5	38.00	44.13	17.22
6	44.24	34.07	21.04
7[b]	58.84	40.51	0

[a] The free fatty acid content resulting from the decomposed soaps was assumed to be 0.65% in each case which is sufficiently accurate for all practical purposes.

[b] Product 7 was interesterified at 250° C. Previous work (3) has shown that use of such a high temperature produces some polymerization compounds.

The peanut oil, which was used in most of the interfacial tension measurements, was prepared in the laboratory from a good grade of crude oil. The oil was heavily alkali-refined and bleached. The last traces of soap were destroyed by washing with 1% hydrochloric acid solution, and the oil was then water-washed until the wash water was neutral to methyl orange. Dispersed water was removed from the oil by deodorizing for 45 minutes at 204° C. (400° F.) under 1 mm. pressure, using the apparatus described by Bailey and Feuge (2). The soybean oil and cottonseed oil were refined, bleached, and purified by exactly the same procedure as was used in the case of the peanut oil.

The interfacial tension measurements were made by the ring method using a Cenco-du Noüy interfacial tensiometer and a platinum-iridium ring of 5.98-cm. mean circumference. The ratio of the mean radius of the ring to the radius of the wire of the ring (R/r) was 53.7. Because some of the solutions would not remain completely liquid at temperatures of 50-60° C.,

---

[1] Presented at the 20th Annual Fall Meeting of the American Oil Chemists' Society, Chicago, Illinois, Oct. 30-Nov. 1, 1946.

[2] One of the laboratories of the Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.

BRIEF FOR APPELLEE

United States Court of Appeals
FOR THE DISTRICT OF COLUMBIA CIRCUIT

Appeal No. 19,275

WOLFGANG BAENITZ AND CHEMISCHE WERKE
WITTEN, APPELLANTS

v.

DAVID L. LADD, COMMISSIONER OF PATENTS, APPELLEE

Appeal from the Judgment of the United States
District Court for the District of Columbia

United States Court of Appeals
for the District of Columbia Circuit

FILED JUL 6 1965

Nathan J. Paulson
CLERK

CLARENCE W. MOORE,
Solicitor, U. S. Patent Office,
Attorney for Appellee

J. F. NAKAMURA,
Of Counsel

Appeal No. 19,275

STATEMENT OF QUESTIONS PRESENTED

In the opinion of the appellee, the questions presented are:

(1) Was the District Court clearly wrong in holding, on the evidence before it, that appellants' patent application claims 25 to 30 do not define patentable invention over the disclosures of Christensen patent No. 2,022,494, Feuge et al. patent No. 2,745,749, Millard's "Physical Chemistry for Colleges", Bailey's "Melting and Solidification of Fats" and Bailey's "Industrial Oil and Fat Product"?

(2) Was the District Court clearly wrong in assuming the use of 80% crude glycerine in the Christensen patent where the existence of 80% crude glycerine was shown by pages 636 and 637 of Bailey's Industrial Oil and Fat Products, which pages were attached to the Brief for Defendant but not introduced at trial?

INDEX

Page

Introduction 1

Counter-Statement of the Case 2

Appellants' application 2
The prior art 3
The claims on appeal 5
The grounds for refusal of the claims 6
Plaintiffs' Exhibits 3, 4, 6 and 7 as shown in the Joint Appendix 6

The Statute involved 7

Summary of argument 7

Argument 9

Appellants' burden of proof 9
Unpatentability over the prior art 9
Allegation of error in denying plaintiffs' motion to vacate judgment 19
The rejection based upon the Kuhrt patent 20
The burden of proof before the District Court 21

Conclusion 22

AUTHORITIES CITED

Cases:

Abbott et al. v. *Coe*, 71 App. D.C. 195, 109 F. 2d 449 21
Field, In re, 161 F.2d 372, 34 CCPA 1066 13
**Johns-Manville Corp.* v. *Ladd*, 117 U.S. App. D.C. 262, 328 F. 2d 563 9
**Shapleigh et al.* v. *Mier*, 299 U.S. 468 14
Slayter, In re, 276 F. 2d 408, 47 CCPA 849 13
Stinson, In re, 154 F. 2d 121, 33 CCPA 853 13
**Urbanic et al., In re*, 319 F.2d 267, 50 CCPA 1564.. 13
**Western States Mach. Co.* v. *S. S. Hepworth Co.*, 147 F.2d 345 (CCA 2, 1945) 13

---

* Cases or authorities chiefly relied upon are marked by asterisks.

United States Court of Appeals
FOR THE DISTRICT OF COLUMBIA CIRCUIT

Appeal No. 19,275

WOLFGANG BAENITZ AND CHEMISCHE WERKE
WITTEN, APPELLANTS

v.

DAVID L. LADD, COMMISSIONER OF PATENTS, APPELLEE

**Appeal from the Judgment of the United States
District Court for the District of Columbia**

BRIEF FOR APPELLEE

INTRODUCTION

Appellants appeal from a decision (J.A. 29) of the United States District Court for the District of Columbia dismissing the complaint (J.A. 1) in an action brought under Section 145 of Title 35, United States Code (Act of July 19, 1952, C. 950, 66 Stat. 803). In the action, the appellants Wolfgang Baenitz, as the applicant, and Chemische Werke Witten, as the assignee, in the Baenitz application, Serial No. 792,706, sought to have the District Court authorize the issuance of a patent containing claims 25 through 30 (J.A. 2-4) of the application. The District Court and the Board of Appeals refused to allow the claims for the reason that they fail to define patentable invention over the prior art under 35 U.S.C. 103.

COUNTERSTATEMENT OF THE CASE

Since the "Statement of Case" in the brief for appellants (pages 2-10) omits essential facts material to the consideration of the questions presented, a counterstatement of the essential facts is deemed necessary.

APPELLANTS' APPLICATION

Appellants' application (J.A. 43-47) relates to a process for producing hard and brittle fats, suitable for food and pharmaceutical purposes, by esterifying a saturated fatty acid having between 10 and 18 carbon atoms per molecule, such as stearic acid, or a mixture of such acids, with an excess of glycerine in the presence of an esterification catalyst. The excess of glycerine is used to control the melting point of the resultant product. The application points out (J.A. 45, fourth paragraph) that the melting point depends upon the amount of excess glycerine used, and specifies (J.A. 44, lines 19-23) the proportion of glycerine to be that amount which produces an ester product having a hydroxyl value of between 40 and 100.

THE PRIOR ART

The Christensen patent No. 2,022,494 (J.A. 53) discloses a process for producing edible fats by esterifying 120 parts by weight of glycerine with from 300 to 600 parts by weight of stearic acid in the presence of a catalyst. Christensen points out (J.A. 54, column 2, lines 31-48) that a product containing 93% monostearin is obtained by using 300 parts by weight of stearic acid, and that a product "consisting mostly of distearin" can be obtained by using 600 parts by weight of stearic acid. He states, further, that ". . . various mixtures of mono and distearin can be prepared by varying the quantity of stearic acid used, in other words by varying the ratio of glycerine to higher fatty acids." Claims 8 and 10 of the patent particularly point out:

> 8. The process of preparing monostearin * * *, the stearic acid being in about the quantity necessary to esterify one of the hydroxyl groups of glycerine.
>
> 10. The process of preparing distearin * * *, the stearic acid being in about the quantity necessary to esterify two of the hydroxyl groups of the glycerine.

The Feuge et al. patent No. 2,745,749 (J.A. 56) describes a process for modifying the melting point and like properties of monostearin by directly acylating a portion of the hydroxyl groups of monostearin (J.A. 56, column 1, lines 54-65). The process comprises acylating (J.A. 56, column 2, line 70 to J.A. 57, column 3, line 7 and column 3, lines 18-22 and 41-46) monostearin with 0.5, 1.0 and 2.0 moles of acetic anhydride per hydroxyl equivalent of monostearin to obtain various mixtures of monostearin and acetylated monostearin. Feuge et al. show (J.A. 58, Table III) that the amount of acetic anhydride used determines the hydroxyl value of the mixtures produced and controls the melting point of the mixtures. Specifically, Table III shows that the melting range and hydroxyl value of products, A-1, A-2 and A-3, obtained from monostearin, are lower, the greater the amount of acetic anhydride used in proportion to the amount of monostearin (J.A. 57, Table I).

The Kuhrt patent No. 2,634,278 (J.A. 60) discloses (J.A. 61, column 1, lines 44-52) the direct esterification of fatty material, such as stearic acid (J.A. 63, column 6, lines 53-57), with an excess of glycerine to produce ester mixtures containing a maximum amount of monoglyceride or a maximum amount of diglyceride (J.A. 64, column 7, lines 55-59). Kuhrt recognizes (J.A. 62, column 3, lines 19-41) that a partial ester product will be in dynamic equilibrium with glycerine and the fatty reactant, and that a change in the relative amounts of glycerine and the fatty reactant will change the character of the product. To carry out the esterification as a cyclic process without reversion of the product, Kuhrt separates the partial ester from the reaction mixture by a rapid vacuum distillation (J.A. 62, column 4, lines 7-24).

The textbook entitled "Physical Chemistry for Colleges," by E. B. Millard, First Edition, published in 1921 (J.A. 71), discloses on pages 93-94 (J.A. 72-73) that the melting point of a substance is lowered by the presence of some other substance in proportion to the quantity of it present.

The textbook, "Melting and Solidification of Fats", by Alton E. Bailey, published in 1950 (J.A. 74), contains on page 48 (J.A. 75) the following statement:

> It is well known that the melting point of a substance is usually lowered by the admixture of a small proportion of any second substance, even if the latter is relatively high melting.

The textbook, "Industrial Oil and Fat Products", by Alton E. Bailey, published in 1945 (J.A. 76), on page 48 (J.A. 77) defines "hydroxyl number" as follows (emphasis quoted):

> * * * The *hydroxyl number* is the number of milligrams of potassium hydroxide equivalent to the hydroxyl content of one gram of fat.

THE CLAIMS ON APPEAL

Claims numbered 25 through 30, found in the Joint Appendix on pages 2-4, are on appeal. The claims are in method form, and claim 25 is quoted by the Board of Appeals (J.A. 48-49) and the District Court (J.A. 30) as a representative claim. Claim 25 defines a method for making a hard and brittle fat in which a fatty acid selected from the group consisting of saturated fatty acids having about 10 to 18 carbon atoms per molecule, and mixtures thereof, is esterified with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, and the melting point of the ester mixture is controlled by selecting the quantity of excess glycerol used. Stearic acid is specified by name in claims 26, 28 and 30 as one of the fatty acids. Claim 27 specifies an excess of glycerol that is "predetermined," and claim 29 calls for the presence of an esterification catalyst.

THE GROUNDS FOR REFUSAL OF THE CLAIMS

Both the Board of Appeals (J.A. 48-41) and the District Court (J.A. 29-34) found that the claims fail to define patentable invention over the Christensen patent (J.A. 53-55) in view of the Feuge et al. patent (J.A. 56-59). The District Court found (J.A. 29-34) further cumulative evidence of unpatentability in Millard's "Physical Chemistry for Colleges" (J.A. 71-73), Bailey's "Melting and Solidification of Fats" (J.A. 74-75) and Bailey's "Industrial Oil and Fat Products" (J.A. 76-77). The claims were also refused by the Board of Appeals (J.A. 48-51) for lack of patentable invention over the Kuhrt patent (J.A. 60-68) in view of the Feuge et al. patent. The District Court, however, stated (J.A. 34), "Since the Court agrees with the Patent Office rejection on the above discussed theory [based on the Christensen patent], it does not find it necessary to reach the rejection based upon the Kuhrt disclosure."

PLAINTIFFS' EXHIBITS 3, 4, 6 AND 7 AS SHOWN IN THE JOINT APPENDIX

The attention of the Court is called to pages 52a-52d of the Joint Appendix which purport to show

Plaintiffs' Exhibits 3, 4, 6 and 7. Those pages are not identical copies of the original exhibits constituting part of the record on appeal, and were printed without the knowledge or consent of the appellee. It is respectfully requested that pages 52a-52d of the Joint Appendix be disregarded by the Court, and that the Plaintiff's Exhibits 3, 4, 6 and 7 in the record on appeal be considered in lieu thereof.

THE STATUTE INVOLVED

The section of the patent statute involved in this appeal is Section 103 of Title 35, United States Code. Section 103 is set out in the brief for appellants on page 11.

SUMMARY OF ARGUMENT

Appellants have the burden of showing that the District Court was clearly wrong in finding that the claims do not define patentable invention. Appellants have failed to make such a showing here.

The Christensen patent shows that it is old in the art of esterifying stearic acid with glycerine to vary the ratio of glycerine to stearic acid in order to control the composition of the ester mixture produced. Claims 8 and 10 of the patent show that the amounts of glycerine used range from 160 to 320 grams per 1000 grams of stearic acid and thus overlap the range of 131 to 168 grams of glycerine per 1000 grams of stearic acid, shown to be used in appellants' process. This overlap is shown to be in the range of ester products having hydroxyl numbers between 90 and 100.

While the illustrative examples described by Christensen reveal the use of 200 to 400 grams of glycerine per 1000 grams of stearic acid, claims 8 and 10 of the patent show that those amounts of glycerine must be assumed to be equivalent amounts of 80% crude glycerine.

The existence of 80% crude glycerine is a fact of which judicial notice can be taken.

The processes of Christensen and appellants appear to be essentially the same and to produce inherently the same result. The advantage of controlling the melting point by the ratio of glycerine to stearic acid used is the only feature of the claimed process which is not described by Christensen. Since the Feuge et al. patent shows that in the art of producing fats by esterification, varying the ratio of reactants was known to control the melting point of the ester mixture obtained, it would have been obvious to one skilled in the art that the ratio of glycerine to stearic acid in the Christensen process controls the melting point of the ester mixture produced. Further evidence of obviousness of that advantage is found in Millard's "Physical Chemistry for Colleges" and Bailey's "Melting and Solidification of Fats".

The District Court correctly denied appellants' motion to vacate judgment. Appellants refer to evidence which assertedly could have been brought forward to show that the District Court could not properly assume that Christensen used 80% crude glycerine. No reason is apparent from that evidence which would preclude the assumption made by the District Court.

ARGUMENT

Appellants' Burden of Proof

In its Opinion (J.A. 29-34), the District Court stated (J.A. 34) that it agreed with the Patent Office rejection. In *Johns-Manville Corp.* v. *Ladd*, 117 U.S. App. D.C. 262, 328 F.2d 563, 140 USPQ 362, the Court said:

> * * * [A]s we have frequently stated, the findings of the Patent Office, an expert administrative body, especially when confirmed by the District Court, will not be overturned here unless clearly infected with error. *Zenith Radio Corporation* v. *Ladd*, 114 U.S. App. D.C. 54, 57, 310 F.2d 859, 862 (1962).

In the same *Johns-Manville* decision, this Court also stated:

> In assaying the action of the district judge, we note also that his findings "shall not be set aside unless clearly erroneous," particularly where those findings, as here, to some extent at least, are predicated on the trial court's judgement of the credibility of the witnesses appearing before it. Rule 52(a), F.R. Civ. P.

Appellants accordingly have the burden of proving that there is no rational basis in the evidence before the District Court for its decision when it agreed with the Patent Office tribunals.

Unpatentability Over the Prior Art

Claim 25, quoted as representative by the Board of Appeals (J.A. 48-49) and the District Court (J.A.

30), defines a method for making hard and brittle fats in which a fatty acid selected from the group consisting of saturated fatty acids having about 10 to 18 carbon atoms per molecule, and mixtures thereof, is esterified with an excess of glycerol to produce an ester mixture having a hydroxyl number between about 40 and 100, and the melting point of the ester mixture is controlled by selecting the quantity of excess glycerine used. Stearic acid is specified by name in claims 26, 28 and 30 (J.A. 2-4) as one of the group of fatty acids, and is identified as the C_{18} fatty acid by the witness Baenitz (J.A. 16 under "Cross Examination").

The appellant Baenitz gave evidence of the actual amounts of glycerine which will give hydroxyl numbers between 40 and 100 when he introduced Plaintiff's Exhibit 6 and testified (J.A. 15, second and third questions) that the amounts of glycerine fall between the lines indicated for hydroxyl numbers 40 and 100. From mere inspection of Plaintiff's Exhibit 6,[1] it is apparent that for the C_{18} (stearic) acid, the actual amounts of glycerine fall within the range between 131 and 168 grams of glycerine per 1000 grams of stearic acid.

Christensen's patent (J.A. 53-55) particularly points out his invention in the following terms in patent claims 8 and 10 (J.A. 55, column 2, lines 31-46):

> 8. The process of preparing monostearin which comprises heating to a temperature of

---

[1] It is respectfully requested that the Court refer to the original Plaintiffs' Exhibit 6.

> about 200-220° C. a mixture of glycerine, stearic acid and a catalyst comprising an alkali-forming metal glycerate, *the stearic acid being in about the quantity necessary to esterify one of the hydroxyl groups of the glycerine.* (emphasis added).
>
> 10. The process of preparing distearin which comprises heating to a temperature of about 200-220° C. a mixture of glycerine, stearic acid and a catalyst comprising an alkali-forming metal glycerate, *the stearic acid being in about the quantity necessary to esterify two of the hydroxyl groups of the glycerine.* (emphasis added).

In describing the claimed processes, Christensen discloses (J.A. 54, column 2, lines 27-29 and 45-48) that various mixtures of mono- and distearin can be prepared by varying the quantity of fatty acid reacted with the glycerine.

The quantity of stearic acid necessary to esterify one of the hydroxyl groups of glycerine, as pointed out by the witness Morrell (J.A. 21, last question and answer at J.A. 22), is 284 parts by weight (1 mole) of stearic acid to 92 parts by weight (1 mole) of glycerine. Stated otherwise, the quantity of glycerine is $\frac{92}{284}$ or 0.32 grams of glycerine per gram of stearic acid. The quantity of stearic acid necessary to esterify two of the hydroxyl groups of glycerin is 2 x 284 grams of stearic acid to 92 grams of glycerine. Stated otherwise, the quantity of glycerine is $\frac{92}{2 \times 284}$ or 0.16 grams of glycerine per gram of stearic acid. It is thus apparent that claims 8 and 10 of Christensen particularly point out that the amount of glycerine ranges from 160 to 320 grams per 1000 grams of stearic acid in the Christensen process. Christensen's range thus overlaps appellants' range of 131 to 168 grams of glycerine per 1000 grams of stearic acid.

From Plaintiffs' Exhibit 6 it can be seen that the overlap is in the range of ester products having hydroxyl numbers between 90 and 100. The witness Baenitz testified (J.A. 14, last four questions) that the blue and green lines on Plaintiffs' Exhibit 6 represent the theoretical quantities of glycerin necessary per 1000 grams of fatty acid to produce the monoglyceride and diglyceride respectively of the fatty acid. From mere inspection it is apparent that Christensen's range of 160 to 320 grams of glycerine per 1000 grams of stearic (C_{18}) acid coincides with the range embraced by the green and blue lines. It is further clearly apparent, and admitted by the witness Baenitz (J.A. 17, first question), that this range overlaps the range in which ester products having hydroxyl numbers between 40 and 100 are obtained, specifically in the range of hydroxyl numbers between 90 and 100.

Appellants contend (Br. 19 and 24) that Christensen teaches the use of 200 to 400 grams of glycerine per 1000 grams of stearic acid, and therefore there is no overlap with their range of 131 to 168 grams of glycerine per 1000 grams of stearic acid. Appellants base their contention (Br. 19) on the amounts of glycerine shown in illustrative examples described in the Christensen specification on page 2, right column, lines 1-29 (J.A. 54). The claims of a prior art patent, however, are a part of its disclosure. *In re Urbanic et al.*, 319 F.2d 267, 50 CCPA 1564; *In re Slayter*, 276 F.2d 408, 47 CCPA 849; *In re Field*, 161 F.2d 372, 34 CCPA 1066; *In re Stinson*, 154 F.2d 121, 33 CCPA 853.

As above noted, claims 8 and 10 of Christensen particularly point out that the amount of glycerine ranges from 160 to 320 grams per 1000 grams of stearic acid. The Christensen claims are presumed to be valid and therefore to particularly point out his invention, as required by R.S. 4888, the predecessor statute of 35 U.S.C. 112. See *Western States Mach. Co.* v. *S.S. Hepworth Co.* (CCA 2, 1945), 147 F.2d 345, cert. denied 325 U.S. 873.

Claims 8 and 10 of Christensen thus show that in ~~that~~ the illustrative examples described in the specification on page 2, column 2, lines 1-48 (J.A. 54), the 200 to 400 grams of glycerine used per 1,000 grams of stearic acid are equivalent amounts of 80% crude glycerine. Certainly, the amounts of stearic acid necessary to esterify one, or two, of the hydroxyl groups of glycerine, as specified in the claims of Christensen, could not be less than 1, or 2, moles per mole of glycerin. But such would be the case if the 200 or 400 grams of glycerine were assumed to be 100% glycerine.

Appellants contend (Br. 16-18) that since the only suggestion of the existence of 80% crude glycerine is in pages 636 and 637 of Bailey's "Industrial Oil and Fat Products," which were attached to the Brief for Defendant but not introduced at trial, the District Court relied upon evidence not properly before the Court in assuming that 80% crude glycerine was used by Christensen. It should, however, be noted that the opinion of the District Court does not state that Bailey's "Industrial Oil and Fat Products" was relied on as evidence of the existence of 80% crude glycerine. The fact of the existence of 80% crude glycerine is by no means a matter of specialized chemical knowledge. Both the Encyclopedia Brittanica (1957) and the Encyclopedia Americana (1927 and 1964) disclose the existence of 80% crude glycerine. It is suggested that the District Court can be assumed to have taken judicial notice of these well known and readily available general reference works. See *Shapleigh et al.* v. *Mier*, 299 U.S. 468. It should also be noted that claims 8 and 10 of the Christensen patent make it unmistakeably clear that glycerine of 80% strength must be assumed to have been used in the illustrative examples.

Appellants' application specification discloses (J.A. 46, midpage) that their reaction temperature of 210°C. falls within Christensen's range of 200-220°C. There is no suggestion in the specification that the catalyst is a critical factor.

Accordingly, it is submitted that the District Court correctly found (J.A. 33):

> The testimony demonstrated that the amount of glycerine used in the plaintiffs' process ranges from 131 to 168 grams per 1000 grams of stearic acid, and that this clearly overlaps the Christensen range of 160 to 320 grams per 1000 grams, assuming that 80% crude glycerine was used in the reaction. Further, it was shown that the

> hydroxyl numbers of esters obtained from use of 160 to 168 parts of glycerine would be in the range from 90 to 100. It was also pointed out that the temperature at which the Christensen process is conducted is the same as that for plaintiffs' process. From this, it appears that the processes of Christensen and the plaintiffs are essentially the same.

Appellants urge (Br. 27) that their process produces mixtures of triglycerides, diglycerides and small amounts of monoglycerides, that the products of Christensen's process are inherently different and therefore, that the processes are not the same. While the witness, Dr. Morrell, adopted the sweeping conclusion (J.A. 20, 21 and 26) that Christensen teaches the production of only monoglycerides, diglycerides or mixtures of mono- and diglycerides, it is important to note that nowhere in the patent does Christensen state that *only* monostearin, *only* distearin or mixtures of *only* mono- and distearin are produced, or that no tristearin is formed. Only the monostearin products were actually made by Christensen. One is described (J.A. 54, second column, lines 19-24) as showing a yield of 96-97% of the theoretical for monostearin, and the other is described (J.A. 54, second column, lines 40-42) as showing a yield of monostearin of about 93% of the theoretical. Of the distearin products, Christensen's only description (J.A. 54, second column, lines 42-48) is of a "product consisting mostly of distearin." It is, thus, apparent that Christensen's products are mixtures. While those mixtures are not completely characterized by Christensen, there is no evidence, apart from the testimony of Dr. Morrell, that Christensen's products, particularly the "product consisting mostly of distearin," are in fact different from the products of appellants' process.

Accordingly, it is submitted that the District Court, on the record before it, correctly found (J.A. 33):

> Accordingly, it seems reasonable to suppose they [the processes of Christensen and Baenitz] would inherently produce the same result.
>
> At least, the Court cannot say such a view is unwarranted where plaintiffs have offered no convincing proof to the contrary.
>
> On the question of controlling the melting point, it is clear, first, that if Christensen's process inherently produces plaintiffs' result, then the feature of controlling the melting point would necessarily be included in such a process.

As shown by Christensen, it was a known procedure in the art to vary the ratio of glycerine to fatty acids in order to vary the composition of the ester mixture produced. It is apparent, then, that the only recitation in the claims, not met by Christensen, is the reference to the procedure of varying the ratio of glycerine to fatty acid as a procedure for controlling the melting point of the ester mixture produced.

The prior art, as shown by the Feuge et al. patent (J.A. 56-59), shows that the ratio of acetic anhydride to monostearin in a process for producing fats by acylation of monostearin was known to control the melting point of the fat obtained and the amount of monostearin in proportion to acylated monostearins in the fat. As shown by the testimony of the witness, Dr. Morrell (J.A. 26, top half page), acylation of monostearin is an esterification with acetic acid, and an acylated monostearin is an ester. Moreover, both Millard's "Physical Chemistry for Colleges" (J.A. 71-73) and Bailey's" Melting and Solidification of Fats" (J.A. 74-75) show that the melting point of a substance was known to be lowered by the presence of a second substance, and in proportion to the quantity of the second substance, as further pointed out by Bailey. It is submitted that it would be wholly obvious to one skilled in the art that selecting the ratio of glycerine to fatty acid used in the Christensen process controls the melting point of the ester mixture produced.

Appellants point out (Br. 30-31) that the Feuge et al. patent is concerned with changing the characteristics of an end product of the Baenitz process, and that the acylated products of Feuge et al. are flexible and stretchable, and not hard and brittle as claimed. It should, however, be noted that the patent clearly teaches the control of melting point of a fat through an esterification reaction by varying the ratio of the reactants, and clearly shows a direct relationship between the melting point and composition of the fat obtained. This teaching is so analogous to appellants' proceess that the differences pointed out by appellants would seem to be without significance.

Appellants further urge (Br. 29-30) that fats produced by their process have a minimum melting point within the range of hydroxyl numbers from 40 to 100, as shown by the V shape of curves on Plaintiffs' Exhibits 4 and 7, and that (Br. 31) Feuge et al. indicate no criticality in melting points for the claimed hydroxyl number range of 40 to 100. While appellants admit (Br. 31) that Feuge et al. teach an increase in melting points with an increase in hydroxyl number of the product, appellants urge that the melting points for the Baenitz products *decrease* with increase of hydroxyl number in the claimed range. It is apparent, however, that Plaintiffs' Exhibit 4 and 7[2] show only an *increase* in melting points of the stearic acid esters (green curve No. 3) with an increase in hydroxyl number in the claimed range between "about 40 and 100." It would, thus, seem clear that there is no merit in appellants' assertion of criticality.

Appellants contend (Br. 32) that the admixture of a second substance with another, as taught in Bailey's "Melting and Solidification of Fats," is different from a chemical reaction producing a mixture, as in appellants' process. Since Millard's "Physical Chemistry for Colleges" shows that melting point lowering is not due solely to the admixture of a second substance, it is believed clear that the asserted difference is without significance.

Accordingly, it is submitted that the District Court, on the record before it, correctly found (J.A. 33-34):

> It is clear, however, from the other references, that varying the proportion of ingredients in mixtures such as Christensen's was, at the time of the invention, known to vary the mixture's

---

[2] It is respectfully requested that the Court refer to the original Plaintiffs' Exhibits 4 and 7.

> melting point. This was shown by the various textual publications introduced by the defendant at trial, and also by the patent to Feuge et al.
>
> In view of this, the Court cannot say the Patent Office was clearly wrong in finding that control of the melting point as claimed by plaintiffs, would have been obvious to a person skilled in the art having all these disclosures before him.

Allegation of Error In Denying Plaintiffs' Motion To Vacate Judgment

Appellants assert (Br. 19-20) that an overlap in the amounts of glycerine used in appellants' and Christensen's process does not exist and can be found only if the assumption that 80% crude glycerine was used by Christensen is proper, that pages 636 and 637 of Bailey's "Industrial Oil and Fat Products" would be insufficient basis for the assumption, even if in evidence, and that, therefore, the District Court erred in denying plaintiffs' Motion to Vacate Judgment Under Rule 59 F.R.C.P.

As hereinbefore noted, claims 8 and 10 of the Christensen patent clearly show that there is an overlap in the amounts of glycerine and that the glycerine described in the illustrative examples must be assumed to be of 80% strength. Appellants urge (Br. 21) that the Bailey publication shows that 80% crude glycerine contains impurities, and that the use of Christensen's products in edible materials precludes the assumption that Christensen used 80% crude glycerine. Christensen, however, shows that extreme purity of the products was not a vital requirement. As pointed out by Christensen (J.A. 54, second column, lines 49-56), the products are incorporated in very small amounts in edible oils, etc., and the catalysts used (sodium glycerate or sodium stearate soap) need not be removed. Further, while appellants assert (Br. 22) that it could have been shown that water normally present in 80% crude glycerine would impair the reaction, it would not necessarily follow that 80% crude glycerine could not be used. Finally, appellants have appended to their brief (Appendix 2 and Appendix 3), two publications said to show (Br. 23-24) that substantially pure 100% glycerine is used in preparing products for food or pharmaceutical uses. It is apparent, however, that those publications show the preparation of monoglycerides of high purity for experimental purposes which might or might not be required for the type of use described by Christensen.

It is, accordingly, submitted that no reason is apparent why Christensen cannot be assumed to have used 80% crude glycerine in the illustrative examples, and that the District Court correctly denied (J.A. 41) appellants' motion to vacate the judgment.

The Rejection Based Upon the Kuhrt Patent

The District Court did not pass upon the further finding by the Board of Appeals (J.A. 49) that the claims do not define patentable invention over the Kuhrt patent (J.A. 60-68) in view of the Feuge et al. patent. Accordingly, should the Court reverse the judgment of the District Court, it would appear that a remand of the case to the District Court for a decision on the rejection based upon the Kuhrt patent would be required.

The Burden of Proof Before the District Court

Appellants urge (Br. 33-34) that it was reversible error for the District Court to apply the "clearly erroneous" test in ruling (J.A. 34) on the Patent Office finding of obviousness in controlling the melting point. It is submitted that the adjudications of this Court do not support appellants' position. Thus, in *Abbott et al.* v. *Coe,* 71 App. D.C. 195, 109 F.2d 449, this Court stated:

> While the judgment of Patent Office officials is not absolutely binding on the courts, it is entitled to great weight, *and is to be overcome by clear proof of mistake* (emphasis added).

CONCLUSION

It is respectfully submitted that, for the foregoing reasons, the conclusions reached by the Board of Appeals and the District Court that the claims at bar are not patentable over the Christensen patent and Feuge et al. patent are correct and have a reasonable basis in the record, that the District Court correctly found additional evidence of non-patentability in Millad's "Physical Chemistry for Colleges," Bailey's "Melting and Solidification of Fats" and Bailey's "Industrial Oil and Fat Products," and that under the standard of review reiterated by this Court in *Johns-Manville Corp.* v. *Ladd, supra,* the judgment of the District Court should be affirmed.

Respectfully submitted,

CLARENCE W. MOORE,
Solicitor, U. S. Patent Office,
Attorney for Appellee

J. F. NAKAMURA,
Of Counsel

July 1965

☆ U. S. GOVERNMENT PRINTING OFFICE: 1965 779862 932

REPLY BRIEF FOR APPELLANTS

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 19,275

WOLFGANG BAENITZ, and
CHEMISCHE WERKE WITTEN,
Appellants,

v.

DAVID M. LADD,
Commissioner of Patents,
Appellee

Appeal From the United States District Court
For the District of Columbia

United States Court of Appeals
for the District of Columbia Circuit

FILED SEP 3 1965

Nathan J. Paulson
CLERK

PAUL M. CRAIG, JR.
Pennsylvania Building
Washington, D. C.
Attorney for Appellants

DICKE & CRAIG
Pennsylvania Building
Washington, D. C.
Of Counsel

INDEX

INTRODUCTION 1

ARGUMENTS:

1. Reliance on Improper Evidence 2
2. Applicability of Assumption of 80% Crude Glycerine Use to Christensen 5
3. Identity of Christensen and Baenitz Processes and Products 8
4. Obviousness of Invention 10
5. Burden of Proof 12

GENERAL REMARKS 13

CONCLUSION 17

TABLE OF CASES

Collins v. Owen, 199 F. Supp. 61 8

Decker v. Federal Trade Commission, 84 App. D.C. 137, 176 F.2d 461, cert. denied, 338 U.S. 878 7

Dominion Magnesium Ltd. v. United States, 320 F.2d 388 8

Dotter, Ex parte, 12 U.S.P.Q. 382 9

Genesco, Inc. v. Greeson, 125 S.E. 2d 786 4

Hahn and Clay v. A.O. Smith Corp., 320 F.2d 166 8

Kinnear-Weed Corp. v. Humble Oil and Refining Co., 150 F.Supp. 143 8

Minneapolis-Honeywell Regulator Co. v. Midwestern Instruments, Inc., 148 F.Supp. 248 8

Muench, Ex Parte, 79 U.S.P.Q. 92 9

Oregon Saw Chain Corp. v. McCulloch Motors Corp., 323 F.2d 758 8

Parmelee Pharmaceutical Co. v. Zink, 285 F.2d 465 9

Smith v. Kingsland, 84 App. D.C. 284, 178 F.2d 26 7

Sperry Rand Corp. v. Texas Instruments, Inc., 206 F.Supp. 676 8

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 19,275

WOLFGANG BAENITZ, and
CHEMISCHE WERKE WITTEN,
Appellants,

v.

DAVID M. LADD,
Commissioner of Patents,
Appellee

*Appeal From the United States District Court
For the District of Columbia*

REPLY BRIEF FOR APPELLANTS

The following Reply Brief is respectfully submitted on behalf of appellants pursuant to Rule 17 (c), General Rules of the United States Court of Appeals for the District of Columbia Circuit.

INTRODUCTION

The following arguments are respectfully submitted to point out the certain inaccuracies and fallacies in appellee's arguments as presented in his Brief where not only new arguments have been presented for the first time but decisions have been cited in support of propositions which either have no bearing on these arguments or misconstrue the decisions in the respective cases.

Before dealing at length with appellee's Brief, it may be recalled that the correctness of the decision of the District Court hinges on the identity between the process and resulting products, on the one hand, as taught by the Christensen patent No. 2,022,494 (JA 53) and, on the other, as defined by the claims in the application of appellant Baenitz. As pointed out at length in the Brief for Appellants under the heading of "2. Failure to Grant Motion to Vacate Judgment" and under the heading of "4. Identity of Christensen and Baenitz Processes and Products," neither the formulation, i.e., neither the amounts of ingredients used in the respective processes nor the resulting products are the same. This is evident from the following comparison:

Christensen	Baenitz
Amounts of Glycerine Used for 1,000 Grams of Stearic Acid	
200-400 grams, producing pure monoglycerides, pure diglycerides, or mixtures of pure mono- and diglycerides	131-168 grams, producing mixtures consisting principally of triglycerides and diglycerides with small amounts of monoglycerides
Products Obtained from Process	
either pure monoglycerides, pure diglycerides or mixtures of only pure mono- and diglycerides	mixtures consisting essentially of triglycerides and diglycerides with small amounts of monoglycerides

ARGUMENTS

1. Reliance on Improper Evidence

The foregoing clearly indicates that based on the uncontroverted facts of the case, there is no identity between the amounts of starting materials used in the Christensen and Baenitz processes and in the corresponding products obtained therefrom.

Faced with the realities of non-identity owing to the uncontroverted testimony in the lower Court, appellee's only solution lay in finding a correction factor to be applied to the formulation of Christensen to re-establish the requisite identity or overlap. In the absence of any evidence in the record at the completion of the trial to support a correction factor to be applied to Christensen, appellee submitted evidence, not properly before the Court, after trial with his Brief, urging that a correction factor of 80% should be assumed with respect to the type of glycerine used by Christensen. That the entire logic of the lower Court's decision rests on this assumed correction factor is clear from its decision in response to Plaintiffs-Appellants' Motion to Vacate Judgment.

It may be also recalled that the 80% correction factor was drawn out of thin air, so to speak, from a description of the recovery of glycerol, submitted to the lower Court after trial by appellee in his Brief (reprinted on pages 35 and 36 of appellants' Brief), in which the various steps for the recovery of glycerol are described, these steps consisting of the intermediate step of producing at first the "half-crude glycerine" with a glycerol concentration of about 40%, the next intermediate step of further concentrating the "half-crude glycerine" to 80% crude glycerine and the final step of converting the 80% crude glycerine into the commercial grades of glycerines as normally used. Realizing that reliance by the lower Court on such improperly submitted evidence is normally reversible error, appellee now, for the first time, suggests that the assumption made by the Court as to the use of 80% crude glycerine by Christensen is a fact of which judicial notice can be taken.

At the outset, it should be noted that the lower Court's opinion is devoid of any indication of judicial notice, nor was this doctrine ever urged before by appellee.

Furthermore, this is indeed a new and startling enlargement of the doctrine of judicial notice to situations where the facts are controverted, could be disproved if properly tried as pointed out on pages 22, 23, and 24 of appellants' Brief, and lack the requisite notoriety to bring the same within the permissive bounds of the legal doctrine invoked by appellee.

By definition, judicial notice is the "cognizance of certain facts which judges and jurors may properly take and act on without proof because they already know them." It is also well known that any matter, to be a subject of judicial notice, must be known and well established and authoritatively settled, not doubtful or uncertain. While the doctrine of judicial notice rests on the wisdom and discretion of the Courts, a prerequisite of notoriety must exist before the doctrine can be invoked.

Stated differently, if witnesses might differ as to scientific principles and deductions to be drawn from circumstances, the subject matter cannot be said to be of such general scientific belief as to justify judicial notice; nor can judicial notice be taken of any scientific fact that is subject to being disproved. *Genesco, Inc. v. Greeson,* 125 S.E.2d 786.

Applying the foregoing to the instant situation, the doctrine of judicial notice cannot be properly invoked merely to escape the consequences of improperly introduced evidence submitted by appellee in an attempt to support his position, which was disproved by the evidence properly adduced at trial. It is, at best, speculative and conjectural whether Christensen intended to use 80% crude glycerine, though even the conjecture is fully refuted by the arguments presented in appellants' Brief. Furthermore, the assumption of 80% crude glycerine is argumentative in nature, as is most evident from appellee's Brief as will appear also more fully hereinafter.

Considering all these factors, there is no place for judicial notice in the instant situation, particularly as it appears to be a mere afterthought on the part of appellee, having never been raised or even suggested before. *Shapleigh et al v. Mier,* 299 U.S. 468, cited on page 14 of appellee's Brief certainly does not support the application of the doctrine of judicial notice to the instant situation. As a matter of fact, in this case judicial notice was assumed though the Court also pointed out that there was no basis for such judicial notice, and even with the assumption of judicial notice, the Supreme Court held against the petitioners in this case.

Accordingly, it is respectfully submitted that assumed judicial notice should not be sanctioned in the instant case merely to escape the consequences of the evidence improperly submitted after trial when its application was urged upon the Court without opportunity of cross examination and/or rebuttal to a set of facts making the application completely argumentative and conjectural rather than of such notoriety as to leave no room for any possible reason of its non-application.

2. Applicability of Assumption of 80% Crude Glycerine Use to Christensen

It is worthy of note that appellee's Brief fails to take issue with the reasons advanced on pages 20 through 24 of appellants' Brief why the assumption of 80% crude glycerine in Christensen is obviously incorrect, especially considering the intended use of the products obtained by Christensen.

Similarly, appellee's Brief is totally silent as to why this assumption is made only with respect to Christensen yet no such assumption is made with respect to the Baenitz process, considering appellee's contention that the processes and products are identical; and both are equally silent as to the type of glycerine used.

Surely appellee would have dealt with these issues with equal thoroughness if a logical basis existed therefor. However, the absence of any arguments in appellee's Brief merely further indicates the genuine lack of existence of reasons and thus the incorrectness of the assumption applied only to Christensen.

To sustain the untenable position, appellee now advances a novel doctrine; namely, that the use by Christensen of 80% crude glycerine must be inferred because the claims fail to specify what type of glycerine was used. The claims of a patent admittedly form part of its disclosure. However, the cases cited at the top of page 13 of appellee's Brief do not support appellee's contention that broad claims should be construed in contradiction to the specific disclosure of the patent to arrive by the use of an arbitrarily chosen correction factor and by modifying the specific disclosure at a desired result.

Looking at the case of *In re Urbanic et al.*, 319 F.2d 267, 50 CCPA 1564, cited by appellee on page 13, the issue decided in this case was that a patent, as a whole, should be considered in connection with the question of patentability. In this case, the prior art patent disclosed a range of from "10 to 55%" of a compound in the specification though in the working examples 45% of the compound was used. The Court held that the disclosure as a whole including the range disclosed in the specification must be considered and not the working examples. If anything, this case holds that the described ranges in the specification of a patent as to the amount of a compound used are to be considered in connection with the question of patentability rather than a specific amount or amounts set forth in the working examples. However, this is a far cry from what appellee is attempting to do in the instant case; namely, to modify the range of amounts of compounds used, as disclosed in the Christensen patent, by referring to broad claim language. In the Christensen patent, the amount of glycerine per 1,000 grams of stearic acid is clearly defined, the range being given on page 2, right column, lines 1 through 29 of the patent (JA 54) where the patentee clearly prescribes the use of 120 parts by weight of glycerine for 300 parts by weight of stearic acid to produce substantially pure monostearin and 120 parts by weight of glycerine for 600 parts by weight of stearic acid to produce pure distearin, these values, as pointed out on page 19 of appellants'

Brief corresponding to 400 grams of glycerine per 1,000 grams of stearic acid for the monostearin and 200 grams of glycerine per 1,000 grams of stearic acid to produce the distearin. Obviously, any mixtures of mono- and distearin fall within the range of 400 to 200 grams of glycerine per 1,000 grams of stearic acid.

The second case of *In re Slayter*, 276 F.2d 408, 47 CCPA 489, cited by appellee on page 13 of his Brief also does not support the contention for which it was cited. In this case the Court merely held that the patent specification (and not the claims) made it clear that the specific article illustrated is only one embodiment of the invention and that there would be no purpose in concentrating the fibers in one part of the glass where the particular reasons applicable to the illustrated embodiment do not exist. However, this decision certainly does not support the proposition that broad claims should be interpreted in contradistinction to the specification and to the specific ranges disclosed.

Both of the other cases cited on page 13 of appellee's Brief are equally deficient and inappropriate to sustain the contention that the broad language of a claim may be resorted to to modify the disclosure found in the specification.

Quite the contrary, it has been repeatedly held that the claims are read in the light of the specification. *Decker v. Federal Trade Commission*, 84 App. D.C. 137, 176 F.2d 461, certiorari denied, 338 U.S. 878. As also pointed out by this Court in *Smith v. Kingsland*, 84 App. D.C. 284, 178 F.2d 26, the claims of a patent must be read in the light of the specification not for purposes of enlarging them, but to ascertain the meaning and intention of the patentee with respect to the claims. In the instant situation, appellee seeks exactly the reverse.

The attention of this Court is also respectfully directed to the following decisions, all of which point out that in order to construe a claim or claims of a patent correctly to determine what was the actual invention disclosed in the description and claimed by the patentee, a Court must read the claims in the light of the description and drawings, if any. *Dominion Magnesium Ltd. v. United States*, (U.S. Ct. of Claims) 320 F.2d 388; *Hahn and Clay v. A. O. Smith Corp.*, (C.A. -5) 320 F.2d 166. That claims cannot be construed in contradiction with the specification is also a well-established principle of patent law. *Minneapolis-Honeywell Regulator Co. v. Midwestern Instruments, Inc.*, 148 F. Supp. 248; *Kinnear-Weed Corp. v. Humble Oil and Refining Co.*, 150 F. Supp. 143.

The Circuit Court of Appeals for the Ninth Circuit has re-affirmed this principle recently in *Oregon Saw Chain Corp. v. McCulloch Motors Corp.*, 323 F.2d 758, where the Court held that

> The claim of a patent is to be understood and interpreted in the light of its specifications. (Citations)

The attention of this Court is also respectfully directed to such decisions as *Collins v. Owen*, 199 F. Supp. 61, *Sperry Rand Corp. v. Texas Instruments, Inc.*, 206 F. Supp. 676, and many others which could be cited to point out that the claims are interpreted in the light of the specification and not vice versa.

Under these circumstances, the attempt to modify the specific disclosure of a patent including the specific range of amounts of compounds to be used by reference to broad claim language is totally unwarranted and in contravention of well-established principles of patent law.

The foregoing clearly indicates that appellee's attempt to justify the assumption of 80% crude glycerine by Christensen is contrary to well-settled principles of law.

3. Identity of Christensen and Baenitz Processes and Products

The foregoing again clearly indicates that in the absence of the erroneous assumption of 80% crude glycerine use by Christensen, the processes of Christensen and of Baenitz are different, ignoring for the moment the omission in Christensen of the feature and advantage of controlling the melting point by the ratio of glycerine to stearic acid, as admitted in the second full paragraph on page 8 of appellee's Brief.

It should come as no surprise that the resulting products from the respective processes are also different. Christensen discloses the production of monoglycerides, diglycerides, and mixtures of only mono- and diglycerides while the uncontroverted evidence established that the mixtures resulting from plaintiffs' process consist of triglycerides and diglycerides with only small amounts of monoglycerides. On page 15 of appellee's Brief, it is admitted that

> Only the monostearin products were actually made by Christensen.

At the bottom of page 15 of appellee's Brief, one finds the remarkable statement

> While those mixtures are not completely characterized by Christensen, there is no evidence, apart from the testimony of Dr. Morrell, that Christensen's products, particularly the 'product consisting mostly of distearin,' are in fact different from the products of appellants' process.

One only needs to look at the Christensen patent, and more particularly to such claims as claims 1 and 13, to find language to the effect of a process

> of preparing fatty acid esters of the class consisting of mono- and diglycerides. (emphasis added)

The words "consisting of" are words of art in patent language and have a definite meaning which is restrictive in nature. When a claim speaks of a mixture consisting of elements A and B, this means that such mixture includes only elements A and B. This is too well established as to require numerous citations. *Ex parte Muench,* (Patent Office Board of Appeals), 79 USPQ 92; *Ex parte Dotter,* (Patent Office Board of Appeals), 12 USPQ 382; *Parmelee Pharmaceutical Co. v. Zink,* (CA-8), 285 F.2d 465.

Thus, there can be no doubt that the disclosure of Christensen reveals only the production of monostearin and distearin and mixtures consisting of only monostearin and distearin. Accordingly, there also can be no question of identity of the products of Christensen and as obtained by the Baenitz process.

4. Obviousness of Invention

Thus, to uphold the lack of patentability of the invention claimed by appellant Baenitz, appellee must first pass the successive hurdles concerning the true disclosure of the Christensen patent which, according to appellee's logic, involves, in the instant situation, (a) assumption of judicial notice of a controverted fact urged upon the Court by reference to evidence submitted after trial, (b) establishment of the correctness of the argumentative application of such fact—of which judicial notice has to be taken—to the teaching of Christensen but non-applicability thereof to Baenitz (though both disclosures are equally silent on this point), (c) proof of why Christensen must be assumed to use 80% crude glycerine when the impurities in the crude glycerine would jeopardize the chemical reactions and render the products unfit for the intended purpose as edible materials, and (d) the correctness of the proposition to modify the specific range of the amounts of glycerine to be used in the Christensen patent, contrary to its very disclosure, by reference to the broad language in the claims.

Yet, assuming that appellee satisfactorily overcomes all of these hurdles, there still remains the question of obviousness under 35 U.S.C. 103 since Christensen, as admitted by the Court in its decision and by appellee in his Brief, did not disclose the feature and attendant advantage of controlling the melting point by using the predetermined ratio of glycerine to fatty acid to produce a hydroxyl number falling within the claimed range of 40 to 100.

At the bottom of page 16, appellee seeks to convey in his Brief the idea that Feuge et al was interested primarily in controlling the melting point of the fat obtained. This is clearly not the case; any changes in the melting point of the reaction products of Feuge et al are merely incidental to the intended change of the physical characteristics of the product resulting from the chemical reaction according to the Feuge et al patent (JA 56-59). As pointed out previously, this patent acylates, with the use of acetic acid, monoglycerides, which are precisely the compounds present in only small quantities in the mixture product obtained by the process according to Baenitz. In other words, the mixture process of Baenitz consisting of mostly tri-, di- and small amounts of monoglycerides would first have to be converted, for example, according to the process of the Kuhrt patent 2,634,278 (JA 60-68) into monoglycerides before the Feuge et al process could be used. Moreover, Feuge et al is concerned with

> chemically modifying certain glyceridic mixtures so that the modified mixture exhibits the property of texture, melting point, flexibility, and the like desired for a particular utilization. (column 1, lines 21 through 25, JA 56). (emphasis added).

Insofar as Table III of Feuge et al is concerned (JA 58), this table merely shows what has been known for some time; namely, that the higher the hydroxyl number, the higher normally the melting point. Looking, for example, at plaintiffs' Exhibit 7 (JA 52d) one may see that for the various esters having between 10 and 18 carbon atoms per molecule, the melting point is normally lowest for the ester having a hydroxyl number of zero and increases with increase in the hydroxyl number. It may be recalled that the points indicated by the small squares, triangles, and circles of plaintiffs' Exhibit 7 (JA 52d) are based on information derived from the prior literature as set forth in plaintiffs' Exhibit 2 (JA 52). However, nowhere does Feuge et al suggest that within the claimed range of 40 to 100 the melting point of a particular reaction mixture is less than the minimum melting point known heretofore in connection with the particular compounds making up the mixture.

Considering further the fact that the claims of the Baenitz application call for hard and brittle fats whereas Feuge et al is concerned with flexible, non-greasy compositions that can be stretched and elongated, lack of obviousness certainly cannot be derived from this patent. As to the additional evidence submitted by defendant-appellee at trial (JA 72-75), Millard's *Physical Chemistry for Colleges* (JA 72-73) merely points out what is well known in chemistry; namely, that the purity of a product is determined by the melting point, impurities normally lowering the melting point. However, as is clear from plaintiffs' Exhibit 3 (JA 52a), within the claimed hydroxyl range of 40 to 100, there is no question of purity or impurities but instead reaction mixtures are involved which represent a complicated system containing three (3) components of which two; namely, diglycerides and triglycerides, appear in substantial, almost equal amounts. Furthermore, as pointed out previously, there is a difference between admixture (JA 75) and the presence of a multi-component system in a reaction product as obtained directly by the reaction of the process in accordance with the Baenitz invention.

5. Burden of Proof

On page 9 of appellee's Brief, appellee contends that appellants

> have the burden of proving that there is no rational basis in the evidence before the District Court for its decision when it agreed with the Patent Office tribunals.

Since the decision of the District Court is not in accordance with the evidence but is clearly contrary thereto, this burden is believed clearly met by appellants. However, the quantum of burden of proof according to this dictum is quite different from the "clearly erroneous" test applied by the lower Court in this case.

The foregoing clearly indicates the reversible error in the decision by the lower Court.

General Remarks

A few additional general remarks are submitted in connection with some of the more glaring, incorrect or unsupported statements found in appellee's Brief which are either incorrect or unsupported by the evidence.

At the bottom of page 3 appellee states that Feuge et al (JA 56) "describes a process for modifying the melting point and like properties of monostearin." The physical properties such as stretching, elongation, etc. are not believed to be like properties, particularly as the changes in melting point are only incidental to the changes in these desired properties; namely, the change from a hard and brittle product (Baenitz) into a flexible and stretchable product (Feuge et al).

Dealing with the Kuhrt patent (JA 60) appellee states on page 4 that this patent discloses the direct esterification of fatty material to produce

> ester mixtures containing a maximum amount of monoglyceride or a maximum amount of diglyceride (JA 64, column 7, lines 55-59).

Reference to the text of a patent clearly indicates that it is not maximum amounts but optimum amounts that Kuhrt seeks to produce.

On page 7, appellee's Brief incorrectly states that

> claims 8 and 10 of the patent show that the amounts of glycerine used range from 160 to 320 grams per 1,000 grams of stearic acid and thus overlap the range of 131 to 168 grams of glycerine per 1,000 grams of stearic acid.

There is nothing in claims 8 and 10 which show that the range is 160 to 320 rather than 200 to 400 as specifically disclosed in this patent.

On page 8 of appellee's Brief one finds the startling assertion that in Christensen the

> amounts of glycerine must be assumed to be equivalent amounts of 80% crude glycerine.

Nowhere is there any basis for such required or even possible assumption.

Later on, on page 8, appellee states

> the processes of Christensen and appellants appear to be essentially the same and to produce inherently the same result. The advantage of controlling the melting point by the ratio of glycerine to stearic acid used is the only feature of the claimed process which is not described by Christensen.

This generalization completely overlooks the fact that Christensen gives no indication of hydroxyl numbers nor shows or indicates any correlation thereof with the melting points, not to mention a suggestion of the criticality of the particular melting point to produce the depressing effect as disclosed by Baenitz.

In the last paragraph on page 8 of appellee's Brief, reference is made to the evidence which assertedly could have been brought forward, indicating at the same time that no reason is apparent from that evidence which would preclude the assumption made by the District Court. Such "asserted" evidence speaks for itself, and is believed to clearly indicate that the assumption of 80% crude glycerine in Christensen is controversial, to say the least.

On pages 11 and 12, appellee's Brief speaks about quantities necessary to esterify hydroxyl groups of glycerine. However, these quantities are theoretical quantities as pointed out previously which have no bearing to the realities encountered in practice, as also clearly established by the witnesses at trial.

Turning to the second full paragraph on page 13 of appellee's Brief, it might be mentioned again that the arguments made therein are based on theoretical amounts which have no bearing in practice.

On page 18, in referring to plaintiffs' Exhibits 4 and 7, appellee argues that within the claimed hydroxyl range of 40 to 100, only an increase in the melting points of the stearic acid esters occurs. This, of course, is a fallacious and specious argument since the minimum occurs at a hydroxyl number of about 40 and within the range of 40 to 100, the melting points are below the minimum melting point known heretofore for the respective tri-esters or tri-glycerides.

At the bottom of page 19, appellee states

> Christensen, however, shows that extreme purity of the products was not a vital requirement.

Appellants are unable to find any such showing, a statement merely invented by appellee to support the 80% crude glycerine notwithstanding the use of Christensen's products as edible materials. At the top of page 20, appellee's Brief refers to small amounts incorporated in edible oils. Again, it should be noted that Christensen's catalyst is present in amounts of the order of about 1% (page 3, column 1, line 7, JA 55) whereas under the assumption of 80% crude glycerine, the impurities present in the amount of glycerine used by Christensen would be 20%, i.e., twenty times as much. Moreover, whereas the catalysts are known materials, the impurities would be unknown and probably would be harmful materials as contrasted with the catalysts.

The speculative nature of the entire logic of appellee is best borne out in the sentence found on page 20 of appellee's Brief where it is stated

> it is apparent, however, that those publications show the preparation of monoglycerides of high purity for experimental purposes which might or might not be required for the type of use described by Christensen. (emphasis added)

Appellants have no objection if plaintiffs' Exhibits 3, 4, 6, and 7 in the record on appeal be considered instead of pages 52a and 52d of the

Joint Appendix. Attorneys for appellants wish to reassure this Court that every effort has been made to reproduce these exhibits as accurately as possible notwithstanding the use of multi-color printing and any changes or inaccuracies in the curves are inadvertent. The only changes made were translations of the legends based on the testimony given at trial, and appellants regretfully admit of certain errors. In plaintiffs' Exhibit 3, page 52a, Joint Appendix, in the legend to the left of the Tabulation, "% Free Fat" should read "% Free Fatty Acids."

The legend at the bottom of this Exhibit should read "Glyceride Composition of Esters with Different Hydroxyl Number with Same Fatty Acid Compositions."

In plaintiffs' Exhibit 4, page 52b of the Joint Appendix, the legend for the red curve D should read "coconut oil-refined fatty acid with palm oil fatty acid C_{12}-C_{18} - melting point 45.5°."

Some typographical errors also occurred in the Brief for Appellants, as follows:

On page 30, first full paragraph should read

> The Feuge et al patent (JA 56-59) uses as starting material the partial glyceride mixtures of long-chain saturated fatty acids that may be the final product of the Baenitz process (JA 33).

On page 32 of appellants' Brief, in the first paragraph after the quote, reference is made to the unexpected results in lowering the melting point of the fatty acid mixtures. This should refer to the novel, unexpected results in lowering the melting point of fatty acid glyceride mixtures.

The foregoing errors in the Joint Appendix and in the Brief for Appellants are regretted.

CONCLUSION

It is respectfully submitted that, for the foregoing reasons, the conclusions reached by the District Court as to lack of patentability were clearly erroneous, such conclusions being additionally based on evidence improperly before the Court.

Accordingly, plaintiffs-appellants respectfully submit that the lower Court erred in ordering judgment for the defendant, in denying plaintiffs-appellants' Motion to Vacate the Judgment and in failing to find for plaintiffs-appellants.

Respectfully submitted,

PAUL M. CRAIG, JR.

DICKE & CRAIG
Pennsylvania Building
Washington, D. C.

Attorney for Appellants